

The impact of dynamical screening on the phonon dynamics of LaCuO

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It is shown that dynamical screening of the Coulomb interaction in LaCuO leads to low-energy electronic collective excitations in a small region around the c -axis, strongly mixing with certain interlayer phonons polarized along this axis. The manifestation of such a phonon-plasmon scenario in layered systems based on a nonadiabatic charge response is quantitatively supported by a realistic calculation of the frequency and wavevector dependent irreducible polarization part of the density response function (DRF). The latter is used within linear response theory to calculate the coupled mode dispersion in the main symmetry directions of the Brillouin zone (BZ) and the charge density redistributions excited by certain strongly coupling phonon-like and plasmon-like modes. Moreover, the corresponding mode induced orbital averaged changes of the selfconsistent potential felt by the electrons are assessed. Our analysis should be representative for the optimally to overdoped state of the cuprates where experimental evidence of a coherent three-dimensional Fermi surface and a coherent c -axis charge transport is given. It is demonstrated that modes from the outside of a small region around the c -axis can reliably be calculated within the adiabatic limit. Only a minor nonadiabatic correction is found for these modes in form of stiffening of the high-frequency oxygen-bond-stretching mode at the X-point which is attributed to dynamical reduced nesting. On the other hand, modes inside the nonadiabatic sector of the BZ have to be determined nonadiabatically owing to the poor dynamical screening of the long-ranged Coulomb interaction around the c -axis by the slow charge dynamics. In particular, the relevance of the strongly coupling phonon-like apex oxygen Z-point breathing mode at about 40 meV is emphasized whose mode energy decreases with less doping. Comparing the calculations with experimental neutron scattering and infrared measurements provides strong evidence for low-lying plasmons around the c -axis coupling strongly with corresponding optical phonons. The existence of these coupled modes evidenced by the calculations yields additional virtual exchange bosons for pairing. Finally, the strong nonlocal, nonadiabatic polar electron-phonon coupling around the c -axis found in the computations means that besides the repulsive short-ranged part of the Coulomb interaction responsible for strong correlation effects also the long-ranged part plays an important role for the physics in the cuprates.

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I. INTRODUCTION

In layered, strongly anisotropic materials like the copper oxide high-temperature superconductors (HTSC's) the screening of the Coulomb interaction between the layers is imperfect and its dynamic nature becomes important. Perpendicular to the CuO planes low-lying electronic collective modes (interlayer plasmons) can be expected. In connection with superconductivity the interplay between the attractive interaction, mediated e.g. by phonons, and the dynamically screened Coulomb interaction is more complex than in the conventional theory. Accordingly, the existence and possible relevance of energetically low lying plasmons, e.g. acoustic plasmons, for superconductivity in the cuprates has been discussed before in the literature, see e.g. Refs. 1,2,3,4,5,6. A direct observation of low lying plasmons in the HTSC's, e.g. by high-resolution inelastic electron scattering or some other technique is still missing. As shown in this work there is only a small \mathbf{q} -space region around the c -axis with a nonadiabatic charge response where low lying phonon-plasmon modes can exist and thus a very high \mathbf{q} -space resolution would be

needed in an experiment. However, there is indirect experimental evidence from different probes, like the use of reflectance and ellipsometric measurements⁷, external losses in photoemission⁸, infrared reflectivity⁹ or resonant inelastic x -ray scattering¹⁰.

A proper account of dynamical screening of the Coulomb interaction is not only substantial for superconductivity, but also for a correct description of normal state properties of the HTSC's such as phonon dynamics, see our simplified model approach in Refs. 11,12 and references therein. These calculations indicate that in a small sector around the c -axis of LaCuO phonon-plasmon mixing becomes likely if the interlayer coupling is sufficiently weak. The reason for such a mixing behaviour is a sufficiently slow electron dynamics perpendicular to the CuO planes. However, the important question remains if this is really true in LaCuO or other cuprates. This topic and its consequences is investigated in the present work. Electron dynamics and phonon dynamics then have about the same time scale and as a consequence nonadiabatic behaviour results.

Due to the incomplete dynamical screening of the Coulomb interaction a strong nonlocal, polar electron-

phonon coupling results along the c -axis together with optical conductivity, also in the well-doped metallic state. Experimentally this can be seen for example by the infrared measurements for LaCuO^{13,14,15,16}. Direct evidence for the presence and importance of this type of unconventional electron-phonon coupling around the c -axis where the electrons in the cuprates are strongly coupled to c -axis polar phonons also comes from a recent study of the self-energy of the nodal quasiparticles in Bi2201 (Ref. 17). For a discussion of phonon-plasmon mixing in the cuprates in the context of many-body polaronic effects in the phonon spectrum, we refer to Ref. 18.

As far as underdoped and even optimally doped LaCuO is concerned, the optical c -axis spectra display the features typical for an ionic insulator^{13,14,15,16} and not those of a metal dealt within adiabatic approximation. The spectra are dominated by optical phonons and are almost unchanged from that of the insulating parent compound upon doping. These experimental facts cannot be explained by the first principles calculations of the phonon dynamics of the HTSC's published so far in the literature^{19,20,21,22} because these computations have been performed within (static) density functional theory (DFT) mostly in local density approximation (LDA). Such calculations are based on the adiabatic approximation and always yield a static metallic screening behaviour consistent with closed LO-TO splittings, at the Γ -point, i.e. in particular closed A_{2u} -splittings in LaCuO. This is clearly in contrast to the optical activity in the infrared experiments. In static DFT calculations the transverse effective charges vanish in the metallic state and consequently the induced dipole moments defining the oscillator strengths in the dielectric function (matrix) vanish, too. So, there will be no optical activity by the phonons.

In the first principles calculations^{19,20,21,22} also no explicit results for the phonon dispersion curves along the c -axis or a small region around this axis have been presented. However, this has been accomplished within a simplified model in Refs. 11,12 and will quantitatively be computed in this work. In Ref. 20 only a short remark is given for the case of LaCuO stating that certain experimental results along the c -axis are not supported by the calculation. Moreover, when judging the findings in Refs. 22,23 of a small phonon contribution to the photoemission kink detected by ARPES^{24,25} one should keep in mind that these very interesting calculations also have been performed within static DFT, neglecting e.g. the strong nonadiabatic dynamically screened polar coupling around the c -axis and also correlation effects beyond LDA. On the other hand, the importance of the electron-phonon interaction in context with the kink in the dispersion of the nodal quasiparticles is emphasized in Ref. 26 on the basis of the two-dimensional three-band Hubbard model with electron-phonon interaction included and also in Ref. 27.

Because of the strong anisotropy of the cuprates and consequently an expected very weak dispersion of the

electronic bandstructure along the c -axis, which, however, is systematically overestimated by DFT-LDA calculations, electron dynamics and c -axis phonon dynamics is coupled dynamically and a nonadiabatic treatment with a dynamical screened Coulomb interaction is essential. On the other hand, first principles calculations^{19,20,21,22} and also our microscopic calculations, see e.g. Refs. 12,28,29, have shown that the static DFT is in general a sufficient approximation for phonon modes propagating in the CuO-plane of the HTSC's, but probably not so around the c -axis.

For modes from this \mathbf{q} -space region a coupled phonon-plasmon scenario has been suggested for LaCuO within a simplified microscopic model approach in the framework of linear response theory^{11,30} assuming a sufficiently weak interlayer coupling. In this treatment an eleven-band model (11BM) that represents the two-dimensional electronic structure of the CuO plane in terms of the Cu3d and O_{xy}2p orbitals is generalized to the third dimension by introducing a suitable interlayer coupling in parametrized form. By varying this coupling from the outside it is possible to pass from the strictly two-dimensional nonadiabatic case of the charge response (charge confinement in the CuO plane) to a moderate anisotropic adiabatic situation, typical for DFT-LDA calculations. A comparison with experimental results from inelastic neutron scattering (INS) and infrared spectroscopy also has been given. From the interpretation of the results a phonon-plasmon mixing seems to be likely. Adopting within such a model approach a suitable coupling we were able to explain at least qualitatively an apparent inconsistency between the current interpretation of the INS results for the $\Lambda_1 \sim (0,0,1)$ branches in doped metallic samples of LaCuO (Refs. 31,32). The branches look as expected in adiabatic approximation for a less anisotropic metal, i.e. featuring closed A_{2u} splittings. On the other hand, the infrared data display as already mentioned significant A_{2u} -splittings typical for an ionic insulator, even for well doped metallic probes. Moreover, in Ref. 11 arguments have been presented in the framework of phonon-plasmon mixing to understand at least qualitatively the large softening and the massive line broadening of the apical oxygen breathing mode O_z^Z , at the Z -point of the BZ. Altogether, the model calculations point to phonon-plasmon coupling in a small nonadiabatic region around the c -axis where the Coulomb interaction has to be screened dynamically. A direct experimental search for such a scenario is inconclusive so far^{32,33}.

From our calculations in this work we can conclude that much better \mathbf{q} -space resolution transverse to the c -axis would be needed in the experiment to resolve the coupled mode dispersion in the small nonadiabatic sector.

One important topic of this paper is to scrutinize and quantify the discussion performed within the simplified model for LaCuO by starting with a realistic tight binding approximation (TBA) of the first principles bandstructure (31-band-model (31BM), Ref. 34) which is as

a typical LDA-like bandstructure not anisotropic enough and adapt the latter to the real anisotropy of the material. With such a realistic electronic bandstructure the wavevector and frequency-dependent irreducible polarization part of the DRF is calculated. The latter is used in linear response theory to obtain the dynamically screened Coulomb interaction and ultimately the full nonadiabatic coupled mode dispersion in the main symmetry directions of the BZ. Moreover, charge density redistributions $\delta\rho$ of certain strongly coupling phonon-like and plasmon-like modes of the nonadiabatic region and the corresponding mode induced changes of the self-consistent potential δV felt by the electrons are investigated and compared with the adiabatic limit. From our calculations a phonon-plasmon mixing and a strong nonlocal, nonadiabatic polar electron-phonon interaction is predicted for LaCuO. This conclusion should be representative for the optimally to overdoped state of the cuprates where experimental evidence of a coherent three-dimensional Fermi surface (FS)³⁵ and coherent c -axis charge transport is given^{36,37,38}.

The article is organized as follows. In Sec. II some elements of the theory necessary to understand the calculated results are reviewed. Section III presents the computations. It gives a discussion of the electronic structure and the nonadiabatic and adiabatic mode dispersion. Furthermore, $\delta\rho$ and δV are investigated for certain relevant generic modes in the cuprates. Finally, a summary of the paper is presented in Sec. IV and the conclusions are drawn.

II. ELEMENTS OF THE THEORY AND MODELING

In the following a survey of the theory and modeling is presented. A detailed description can be found in Ref. 39 and in particular in Ref. 40 where the calculation of the coupling parameters of the theory is presented.

The local part of the electronic charge response and the EPI is approximated in the spirit of the quasi-ion approach^{29,41} by an ab initio rigid ion model (RIM) taking into account covalent ion softening in terms of (static) effective ionic charges calculated from a tight-binding analysis. The tight-binding analysis supplies these charges as extracted from the orbital occupation numbers Q_μ of the μ (tight-binding) orbital in question:

$$Q_\mu = \frac{2}{N} \sum_{n\mathbf{k}} |C_{\mu n}(\mathbf{k})|^2. \quad (1)$$

$C_{\mu n}(\mathbf{k})$ stands for the μ -component of the eigenvector of band n at the wavevector \mathbf{k} in the first BZ; the summation in (1) runs over all occupied states and N gives the number of the elementary cells in the (periodic) crystal.

In addition, scaling of the short-ranged part of certain pair potentials between the ions is performed to simulate further covalence effects in the calculation in such a way

that the energy-minimized structure is as close as possible to the experimental one⁴². Structure optimization and energy minimization is very important for a reliable calculation of the phonon dynamics through the dynamical matrix. Taking just the experimental structure data as is done in many cases in the literature may lead to uncontrolled errors in the phonon calculations.

The RIM with the corrections just mentioned then serves as an unbiased reference system for the description of the HTSC's and can be considered as a first approximation for the insulating state of these compounds. Starting with such an unprejudiced rigid reference system non-rigid electronic polarization processes are introduced in form of more or less localized electronic charge-fluctuations (CF's) at the outer shells of the ions. Especially in the metallic state of the HTSC's the latter dominate the *nonlocal* contribution of the electronic density response and the EPI and are particularly important in the CuO planes. In addition, *anisotropic* dipole-fluctuations (DF's) are admitted in our approach^{40,43}, which prove to be specifically of interest for the ions in the ionic layers mediating the dielectric coupling and for the polar modes. Thus, the basic variable of our model is the ionic density which is given in the perturbed state by

$$\rho_\alpha(\mathbf{r}, Q_\lambda, \mathbf{p}_\alpha) = \rho_\alpha^0(r) + \sum_\lambda Q_\lambda \rho_\lambda^{\text{CF}}(r) + \mathbf{p}_\alpha \cdot \hat{\mathbf{r}} \rho_\alpha^{\text{D}}(r). \quad (2)$$

ρ_α^0 is the density of the unperturbed ion, as used in the RIM, localized at the sublattice α of the crystal and moving rigidly with the latter under displacement. The Q_λ and ρ_λ^{CF} describe the amplitudes and the form-factors of the CF's and the last term in Eq. (2) represents the dipolar deformation of an ion α with amplitude (dipole moment) \mathbf{p}_α and a radial density distribution ρ_α^{D} . $\hat{\mathbf{r}}$ denotes the unit vector in the direction of \mathbf{r} . The ρ_λ^{CF} are approximated by a spherical average of the orbital densities of the ionic shells calculated in LDA taking self-interaction effects (SIC) into account. The dipole density ρ_α^{D} is obtained from a modified Sternheimer method in the framework of LDA-SIC⁴⁰. All SIC-calculations are performed for the average spherical shell in the orbital-averaged form according to Perdew and Zunger⁴⁴. For the correlation part of the energy per electron ϵ the parametrization given in Ref. 44 has been used.

The total energy of the crystal is obtained by assuming that the density can be approximated by a superposition of overlapping densities ρ_α . The ρ_α^0 in Eq. (2) are also calculated within LDA-SIC taking environment effects, via a Watson sphere potential and the calculated static effective charges of the ions into account. The Watson sphere method is only used for the oxygen ions and the depth of the Watson sphere potential is set as the Madelung potential at the corresponding site. Such an approximation holds well in the HTSC's^{42,45}. As a general rule, partial covalence reduces the amplitude of the static effective charges in mixed ionic-covalent compounds like the HTSC's, because the charge transfer from

the cations to the anions is not complete as in the entirely ionic case. Finally, applying the pair-potential approximation we get for the total energy:

$$E(R, \zeta) = \sum_{\mathbf{a}, \alpha} E_{\alpha}^{\mathbf{a}}(\zeta) + \frac{1}{2} \sum_{(\mathbf{a}, \alpha) \neq (\mathbf{b}, \beta)} \Phi_{\alpha\beta}(\mathbf{R}_{\beta}^{\mathbf{b}} - \mathbf{R}_{\alpha}^{\mathbf{a}}, \zeta). \quad (3)$$

The energy E depends on both the configuration of the ions $\{R\}$ and the electronic (charge) degrees of freedom (EDF) $\{\zeta\}$ of the charge density, i.e. $\{Q_{\lambda}\}$ and $\{\mathbf{p}_{\alpha}\}$ in Eq. (2). $E_{\alpha}^{\mathbf{a}}$ are the energies of the single ions. \mathbf{a} , \mathbf{b} denote the elementary cells and α , β the corresponding sublattices. The second term in Eq. (3) is the interaction energy of the system, expressed in terms of *anisotropic* pair-interactions $\Phi_{\alpha\beta}$. Both $E_{\alpha}^{\mathbf{a}}$ and $\Phi_{\alpha\beta}$ in general depend upon ζ via ρ_{α} in Eq. (2).

The pair potentials in Eq. (3) can be separated into long-ranged Coulomb contributions and short-ranged terms as follows:

$$\Phi_{\alpha\beta}(\mathbf{R}, \zeta) = \frac{\mathcal{Z}_{\alpha}\mathcal{Z}_{\beta}}{R} - (\mathcal{Z}_{\alpha}\mathbf{p}_{\beta} + \mathcal{Z}_{\beta}\mathbf{p}_{\alpha}) \cdot \frac{\mathbf{R}}{R^3} + \frac{\mathbf{p}_{\alpha} \cdot \mathbf{p}_{\beta}}{R^3} - 3 \frac{(\mathbf{p}_{\alpha} \cdot \mathbf{R})(\mathbf{R} \cdot \mathbf{p}_{\beta})}{R^5} + \tilde{\Phi}_{\alpha\beta}(\mathbf{R}, \zeta), \quad (4)$$

$$\tilde{\Phi}_{\alpha\beta}(\mathbf{R}, \zeta) = K_{\alpha}U_{\beta}(\mathbf{R}, \zeta) + K_{\beta}U_{\alpha}(\mathbf{R}, \zeta) + W_{\alpha\beta}(\mathbf{R}, \zeta) + G_{\alpha\beta}(\mathbf{R}, \zeta). \quad (5)$$

The first term in Eq. (4) describes the long-ranged ion-ion, the second the dipole-ion and the third and fourth term the dipole-dipole interaction. \mathcal{Z}_{α} and \mathcal{Z}_{β} are the variable charges of the ions in case the CF's are excited. The latter reduce to the ionic charges for rigid ions. K_{α} and K_{β} are the charges of the ion cores. The remaining term in Eq. (4) given in (5) represents the short-ranged interactions. These short-ranged contributions to the pair potentials are expressed by the following integrals:

$$U_{\alpha}(\mathbf{R}, \zeta) = - \int d^3r \rho_{\alpha}(\mathbf{r}, \zeta) \times \left(\frac{1}{|\mathbf{r} - \mathbf{R}|} - \frac{1}{R} - \frac{\mathbf{r} \cdot \mathbf{R}}{R^3} \right), \quad (6)$$

$$W_{\alpha\beta}(\mathbf{R}, \zeta) = \int d^3r \int d^3r' [\rho_{\alpha}(\mathbf{r}, \zeta) \rho_{\beta}(\mathbf{r}', \zeta) \times \left(\frac{1}{|\mathbf{r} - \mathbf{r}' - \mathbf{R}|} - \frac{1}{R} - \frac{(\mathbf{r} + \mathbf{r}') \cdot \mathbf{R}}{R^3} \right)], \quad (7)$$

$$G_{\alpha\beta}(\mathbf{R}, \zeta) = \int d^3r [\rho_{\alpha\beta}(\mathbf{r}, \zeta) \epsilon(\rho_{\alpha\beta}(\mathbf{r}, \zeta)) - \rho_{\alpha}(\mathbf{r}, \zeta) \epsilon(\rho_{\alpha}(\mathbf{r}, \zeta)) - \rho_{\beta}(\mathbf{r} - \mathbf{R}, \zeta) \epsilon(\rho_{\beta}(\mathbf{r} - \mathbf{R}, \zeta))], \quad (8)$$

with

$$\rho_{\alpha\beta}(\mathbf{r}, \zeta) = \rho_{\alpha}(\mathbf{r}, \zeta) + \rho_{\beta}(\mathbf{r} - \mathbf{R}, \zeta). \quad (9)$$

$K_{\alpha}U_{\beta}(\mathbf{R}, \zeta)$ yields the short-ranged contribution of the interaction between the core α and the density ρ_{β} according to Eq. (2). $W_{\alpha\beta}(\mathbf{R}, \zeta)$ represents the short-ranged Coulomb contribution of the interaction of the density ρ_{α} with the density ρ_{β} . $G_{\alpha\beta}(\mathbf{R}, \zeta)$ is the sum of the kinetic one-particle- and the exchange-correlation (XC) contribution of the interaction between the two ions⁴⁰. The short-ranged part of the potentials and the various coupling coefficients are calculated numerically for a set of distances R between the ions. The corresponding results are than described by an analytical function of the form:

$$f(R) = \pm \exp\left(\alpha + \beta R + \frac{\gamma}{R}\right). \quad (10)$$

α , β and γ in Eq. (10) are fit parameters. From the adiabatic condition

$$\frac{\partial E(R, \zeta)}{\partial \zeta} = 0 \quad (11)$$

an expression for the atomic force constants, and accordingly the dynamical matrix in harmonic approximation can be derived:

$$t_{ij}^{\alpha\beta}(\mathbf{q}) = \left[t_{ij}^{\alpha\beta}(\mathbf{q}) \right]_{\text{RIM}} - \frac{1}{\sqrt{M_{\alpha}M_{\beta}}} \sum_{\kappa, \kappa'} [B_i^{\kappa\alpha}(\mathbf{q})]^* [C^{-1}(\mathbf{q})]_{\kappa\kappa'} B_j^{\kappa'\beta}(\mathbf{q}). \quad (12)$$

The first term on the right hand side denotes the contribution from the RIM. M_{α} , M_{β} are the masses of the ions and \mathbf{q} is a wave vector from the first BZ.

The quantities $\mathbf{B}(\mathbf{q})$ and $C(\mathbf{q})$ in Eq. (12) represent the Fourier transforms of the electronic coupling coefficients as calculated from the energy in Eq. (3), or the pair potentials in Eqs. (4)-(9), respectively.

$$\mathbf{B}_{\kappa\beta}^{\mathbf{ab}} = \frac{\partial^2 E(R, \zeta)}{\partial \zeta_{\kappa}^{\mathbf{a}} \partial R_{\beta}^{\mathbf{b}}}, \quad (13)$$

$$C_{\kappa\kappa'}^{\mathbf{ab}} = \frac{\partial^2 E(R, \zeta)}{\partial \zeta_{\kappa}^{\mathbf{a}} \partial \zeta_{\kappa'}^{\mathbf{b}}}. \quad (14)$$

κ denotes the EDF (CF and DF in the present model, see Eq. (2)) in an elementary cell. The \mathbf{B} coefficients describe the coupling between the EDF and the displaced ions (bare electron-phonon coupling), and the coefficients C determine the interaction between the EDF. The phonon frequencies $\omega_{\sigma}(\mathbf{q})$ and the corresponding eigenvectors $\mathbf{e}^{\alpha}(\mathbf{q}\sigma)$ of the modes $(\mathbf{q}\sigma)$ are obtained from the secular equation for the dynamical matrix in Eq. (12), i.e.

$$\sum_{\beta, j} t_{ij}^{\alpha\beta}(\mathbf{q}) e_j^{\beta}(\mathbf{q}) = \omega^2(\mathbf{q}) e_i^{\alpha}(\mathbf{q}). \quad (15)$$

The Eqs. (12)-(15) are generally valid and, in particular, are independent of the specific model for the decomposition of the perturbed density in Eq. (2) and the pair approximation Eq. (3) for the energy. The lengthy details of the calculation of the coupling coefficients \mathbf{B} and C cannot be reviewed in this paper. They are presented in Ref. 40. In this context we remark that the coupling matrix $C_{\kappa\kappa'}(\mathbf{q})$ of the EDF-EDF interaction, whose inverse appears in Eq. (12) for the dynamical matrix, can be written in matrix notation as

$$C = \Pi^{-1} + \tilde{V}. \quad (16)$$

Π^{-1} is the inverse of the *irreducible polarization part* of the density response function (matrix) and contains the kinetic part to the interaction C while \tilde{V} embodies the Hartree and exchange-correlation contribution. C^{-1} needed for the dynamical matrix and the EPI is closely related to the (linear) density response function (matrix) and to the inverse dielectric function (matrix) ε^{-1} , respectively.

Only very few attempts have been made to calculate the phonon dispersion and the EPI of the HTSC's using the linear response method in form of density functional perturbation theory (DFPT) within LDA^{19,20,21}. These calculations correspond to calculating Π and \tilde{V} in DFT-LDA and for the *metallic* state only. On the other hand, in our microscopic modeling DFT-LDA-SIC calculations are performed for the various densities in Eq. (2) in order to obtain the coupling coefficients \mathbf{B} and \tilde{V} . Including SIC is particularly important for localized orbitals like Cu3d in the HTSC's. SIC as a correction for a single particle term is not a correlation effect, which per definition cannot be described in a single particle theory, but SIC is important for contracting in particular the localized Cu3d orbitals. Our theoretical results for the phonon dispersion^{12,28,43}, which compare well with the experiments, demonstrate that the approximative calculation of the coupling coefficients in our approach is sufficient, even for the localized Cu3d states. Written in matrix notation we get for the density response matrix the relation

$$C^{-1} = \Pi(1 + \tilde{V}\Pi)^{-1} \equiv \Pi\varepsilon^{-1}, \quad \varepsilon = 1 + \tilde{V}\Pi. \quad (17)$$

The CF-CF submatrix of the matrix Π can approximately be calculated for the metallic (but not for the undoped and underdoped) state of the HTSC's from a TBA of a single particle electronic bandstructure. In this case the electronic polarizability Π in tight-binding representation reads:

$$\begin{aligned} \Pi_{\kappa\kappa'}(\mathbf{q}, \omega = 0) = & -\frac{2}{N} \sum_{n,n',\mathbf{k}} \frac{f_{n'}(\mathbf{k} + \mathbf{q}) - f_n(\mathbf{k})}{E_{n'}(\mathbf{k} + \mathbf{q}) - E_n(\mathbf{k})} \times \\ & \times [C_{\kappa n}^*(\mathbf{k})C_{\kappa n'}(\mathbf{k} + \mathbf{q})][C_{\kappa' n}^*(\mathbf{k})C_{\kappa' n'}(\mathbf{k} + \mathbf{q})]^*. \end{aligned} \quad (18)$$

f , E and C in Eq. (18) are the occupation numbers, the single-particle energies and the expansion coefficients of the Bloch-functions in terms of tight-binding functions.

The self-consistent change of an EDF at an ion induced by a phonon mode $(\mathbf{q}\sigma)$ with frequency $\omega_\sigma(\mathbf{q})$ and eigenvector $\mathbf{e}^\alpha(\mathbf{q}\sigma)$ can be derived in the form

$$\begin{aligned} \delta\zeta_\kappa^\alpha(\mathbf{q}\sigma) &= \left[-\sum_\alpha \mathbf{X}^{\kappa\alpha}(\mathbf{q})\mathbf{u}_\alpha(\mathbf{q}\sigma) \right] e^{i\mathbf{q}\mathbf{R}_\kappa^\alpha} \\ &\equiv \delta\zeta_\kappa(\mathbf{q}\sigma)e^{i\mathbf{q}\mathbf{R}^\alpha}, \end{aligned} \quad (19)$$

with the displacement of the ions

$$\begin{aligned} \mathbf{u}_\alpha(\mathbf{q}\sigma) &= \left(\frac{\hbar}{2M_\alpha\omega_\sigma(\mathbf{q})} \right)^{1/2} \mathbf{e}^\alpha(\mathbf{q}\sigma)e^{i\mathbf{q}\mathbf{R}^\alpha} \\ &\equiv \mathbf{u}_\alpha(\mathbf{q}\sigma)e^{i\mathbf{q}\mathbf{R}^\alpha}. \end{aligned} \quad (20)$$

The self-consistent response per unit displacement of the EDF in Eq. (19) is calculated in linear response theory as:

$$\mathbf{X}(\mathbf{q}) = \Pi(\mathbf{q})\varepsilon^{-1}(\mathbf{q})\mathbf{B}(\mathbf{q}) = C^{-1}(\mathbf{q})\mathbf{B}(\mathbf{q}). \quad (21)$$

A measure of the strength of the EPI for a certain phonon mode $(\mathbf{q}\sigma)$ is provided by the change of the self-consistent potential in the crystal felt by an electron at some space point \mathbf{r} , i.e. $\delta V_{\text{eff}}(\mathbf{r}, \mathbf{q}\sigma)$. Averaging this quantity with the corresponding density form factor $\rho_\kappa(\mathbf{r} - \mathbf{R}_\kappa^\alpha)$ at the EDF located at \mathbf{R}_κ^α , we obtain

$$\delta V_\kappa^\alpha(\mathbf{q}\sigma) = \int dV \rho_\kappa(\mathbf{r} - \mathbf{R}_\kappa^\alpha) \delta V_{\text{eff}}(\mathbf{r}, \mathbf{q}\sigma) \quad (22)$$

as a parameter for the strength of the EPI in the mode $(\mathbf{q}\sigma)$ mediated by the EDF considered. For an expression of $\delta V_\kappa^\alpha(\mathbf{q}\sigma)$ in terms of the coupling coefficients in Eqs. (13) and (14), see Ref. 12.

The generalization for the quantity Π in Eqs. (16) and (17) needed for the kinetic part of the charge response in the nonadiabatic regime, where dynamical screening effects must be considered, can be achieved by adding $(\hbar\omega + i\eta)$ to the differences of the single-particle energies in the denominator of the expression for Π in Eq. (18). Other possible nonadiabatic contributions to C related to dynamical exchange-correlation effects and the phonons themselves are beyond the scope of the present model. Using Eq. (17) for the dielectric matrix, ε , and the frequency-dependent version of the irreducible polarization part, Π , according to Eq. (18), the free-plasmon dispersion is obtained from the condition,

$$\det[\varepsilon_{\kappa\kappa'}(\mathbf{q}, \omega)] = 0. \quad (23)$$

The coupled-mode frequencies of the phonons and the plasmons must be determined self-consistently from the secular equation (15) for the dynamical matrix which now contains the frequency ω implicitly via Π in the response function C^{-1} . Analogously, the dependence on the frequency is transferred to the quantity \mathbf{X} in Eq. (21) and thus to $\delta\zeta_\kappa$ and δV_κ in Eqs. (19) and (22), respectively. Such a nonadiabatic approach is necessary for a description of the interlayer phonons and the charge-response within a small region around the c -axis.

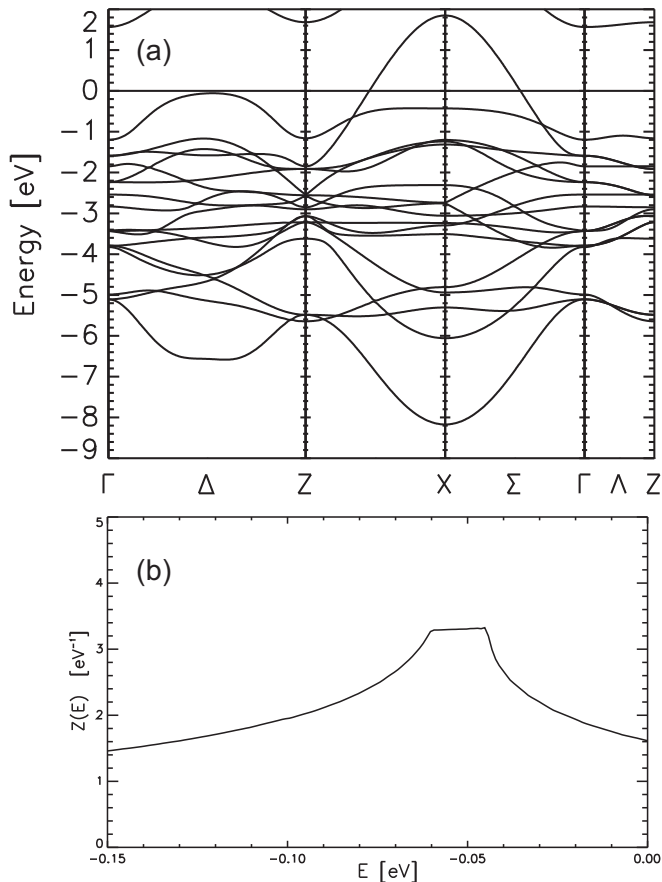


FIG. 1: Electronic bandstructure $E_n(\mathbf{k})$ of LaCuO in the 31 band model (31BM) obtained from an accurate tight-binding representation of the first-principles linearized-augmented-plane-wave bandstructure³⁴. The Fermi energy E_F corresponding to the undoped material is taken as the zero of the energy (a). Density of states $Z(E)$ of the 31BM (b).

The time-consuming numerical calculations were carried out on the computers of the Morfeus GRID at the Westfälische Wilhelms-Universität Münster, with the use of Condor⁴⁶.

III. RESULTS AND DISCUSSION

A. Model of the single particle content of the irreducible polarization part

As a first approximation the electronic bandstructure (BS) $E_n(\mathbf{k})$ of LaCuO and the expansion coefficients $C_{\kappa n}(\mathbf{k})$, needed as input for the single particle content of the irreducible polarization part $\Pi_{\kappa\kappa'}$ in Eq. (18) are taken from an accurate tight-binding representation of the first-principles linearized-augmented-plane-wave bandstructure (LAPW) as obtained within the framework of DFT-LDA³⁴.

This analysis leads to a 31BM including the La5d, Cu3d, 4s, 4p, and O2p states. The result for the re-

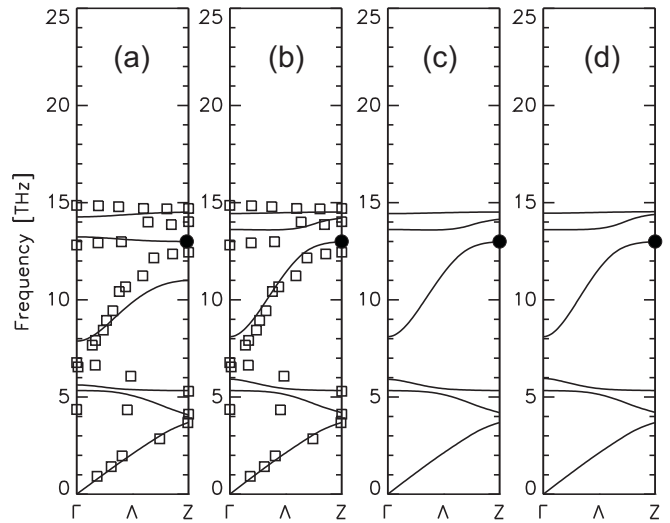


FIG. 2: Calculated phonon dispersion for LaCuO in the adiabatic approximation for the c -axis polarized Λ_1 modes ($\sim (0,0,1)$) based on the bandstructure within the 31BM as input for the proper polarization part $\Pi_{\kappa\kappa'}$ (a). (b)-(d) calculated Λ_1 modes as in (a) but using the modified 31BM (M31BM) as discussed in the text as input for $\Pi_{\kappa\kappa'}$. The panels (b)-(d) represent the phonon dispersion for three different doping levels $x = 0.156$ ($E_F = -0.08$), $x = 0.224$ ($E_F = -0.10$) and $x = 0.297$ ($E_F = -0.135$), by applying a rigid-band approximation of the M31BM to represent the optimally doped to overdoped state of LaCuO. The experimental results (open squares \square) are taken from Refs. 31,32. The full dot (\bullet) denotes the O_z^z mode. E_F is in units of eV.

calculated tight-binding bandstructure is shown in Fig. 1(a) and the corresponding density of states (DOS)

$$Z(E) = \frac{2}{N} \sum_{n\mathbf{k}} \delta(E_n(\mathbf{k}) - E) \quad (24)$$

is displayed in Fig. 1(b). The van Hove peak in the DOS is broadened by the dispersion of the bandstructure in z -direction.

The corresponding calculated phonon dispersion of LaCuO in the adiabatic approximation for the Λ_1 modes polarized along the c -axis being most sensitive with respect to the interlayer coupling is presented in Fig. 2(a). The open squares indicate the present "interpretation" in the literature of the experimental results^{31,32}. This interpretation will be discussed in detail in Sec. III B. The characteristic experimental features of the dispersion are the step-like structure of the second highest branch and most significant the third highest branch with the steep dispersion from the Γ -point towards the Z -point. Both features are not well reflected in the calculation based on the typical DFT-LDA-like bandstructure of the 31BM which underlies the computation of the static ($\omega = 0$) proper polarization part $\Pi_{\kappa\kappa'}$ in Eq. (18). As we see in a moment, the reason for this is an overestimation of the residual k_z dispersion of the bands, i.e. the DFT-LDA-like bandstructure is not anisotropic enough. It should be

remarked that an earlier calculation within the 31BM⁴⁷ neglecting dipole fluctuations as additional electronic polarization processes besides the charge fluctuations still further enhances the deviations between the calculated and the measured result for the Λ_1 -modes. Anisotropic DF's being particular important along the c -axis and for ions in the ionic layers⁴³ have been taken into account in the calculations shown in Fig. 2(a) and in all other computations of the phonon dispersion presented in this paper.

In order to improve the LDA-like electronic BS of the 31BM which overestimates the c -axis coupling we have investigated the effect on the Λ_1 -modes of a modification of the tight-binding parameters of the 31BM. It turns out that a reduction of the first neighbour O_{xy} -La parameters by 1/6 and of the first neighbour La-La parameters by 1/3 leads to a much better result for the calculated Λ_1 -modes in adiabatic approximation, see Fig. 2(b-d). The latter results have been obtained for three different doping levels, where the effect of alloying was treated in rigid-band approximation by lowering the Fermi level appropriately to accommodate x holes per primitive cell.

The modified electronic BS of LaCuO and the corresponding DOS is shown in Fig. 3(a) and Fig. 3(b), respectively. The calculated phonon dispersion is now in good agreement with the experiment. In particular the characteristic features, i.e. the Λ_1 -branch with the steep dispersion and the step-like behaviour are well described. Moreover, we find as a consequence of the enhanced anisotropy a rearrangement of the three Z -point modes with the highest frequencies. While in the 31BM the anomalous O_z^Z mode is the second highest mode in the more anisotropic modified 31BM (M31BM) with a weaker coupling along the c -axis O_z^Z is the lowest of the three modes and the end point of the steep branch.

From a comparison of the BS of the 31BM in Fig. 1(a) with the BS of the M31BM in Fig 3(a) we extract that in the latter case the saddle point region around $\Delta/2$ is more extended and the width of the BS is reduced by about 0.5 eV in the energy range shown in the figure. Important for the charge response along the c -axis is the decrease of the k_z -dispersion of the bands along the Λ direction, which obviously is consistent with the experimental phonon dispersion of the Λ_1 modes being most sensitive to the charge dynamics along the c -axis. Comparing the calculated results for the DOS in both models, see Fig. 1(b) and Fig. 3(b), respectively, we find that the broadening of the van Hove peak is decreased in the M31BM by about 50 % because of the reduced k_z -dispersion. Simultaneously, the DOS $Z(E)$ around the peak is enhanced by the increased anisotropy.

Another important effect of the weakening of the interlayer coupling in our modeling as compared to a typical DFT-LDA bandstructure, leading to a significant enhancement of the DOS $Z(E_F)$ at the Fermi energy E_F , is the amplification of the proper polarization part $\Pi_{\kappa\kappa'}(\mathbf{q})$ on his part. This can be seen in the long wavelength limit

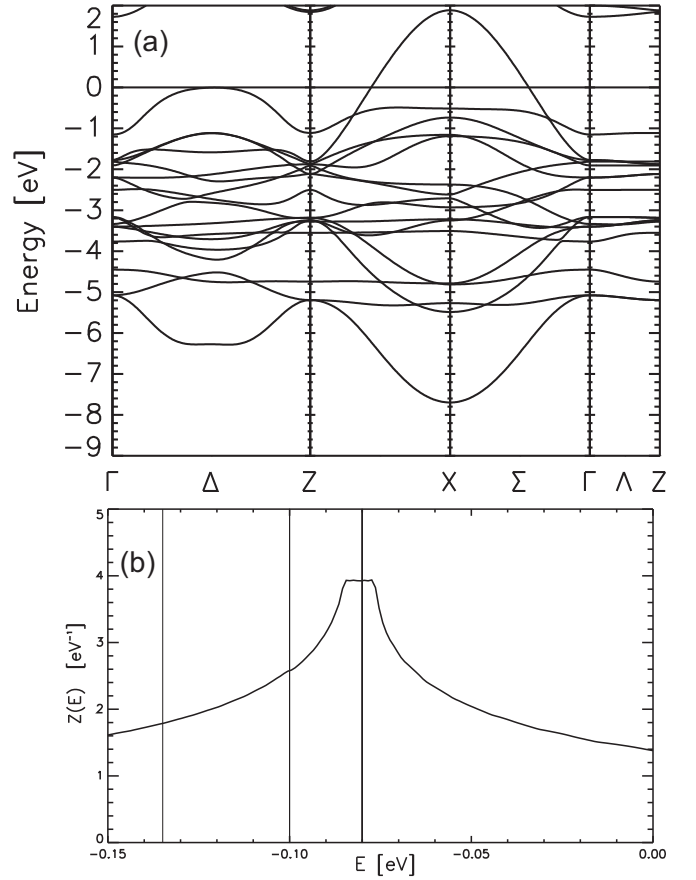


FIG. 3: Electronic bandstructure $E_n(\mathbf{k})$ of LaCuO in the modified 31 band model (M31BM) taking into account the enhanced anisotropy of the real material as compared with the 31BM typical for LDA (a). Density of states $Z(E)$ in the M31BM (b). The vertical lines indicate the different doping levels from optimal to overdoping as in Fig. 2.

where the sum rule

$$\lim_{\mathbf{q} \rightarrow \vec{0}} \sum_{\kappa\kappa'} \Pi_{\kappa\kappa'}(\mathbf{q}) = Z(E_F) \quad (25)$$

rigorously holds for a metal³⁹. Altogether, the results demonstrate the importance of a correct interlayer-coupling also for the electronic properties in the CuO layer.

The vertical lines in Fig. 3(b) indicate three doping levels x in rigid-band approximation. The specific dopings have been selected to model the optimally doped state of LaCuO with the Fermi energy E_F at the van Hove peak (model OP: $E_F = -0.08$ eV, $x = 0.156$) and two overdoped states (model OD1: $E_F = -0.10$ eV, $x = 0.224$; model OD2: $E_F = -0.135$ eV, $x = 0.297$).

The doping levels chosen in this way allow for a comparison of measured Fermi surfaces (FS) at these levels^{48,49} with the calculated FS within the M31BM, see Fig. 4. From the comparison we conclude that the overall features of the BS calculation in the M31BM are in good agreement with the results obtained by angle-resolved

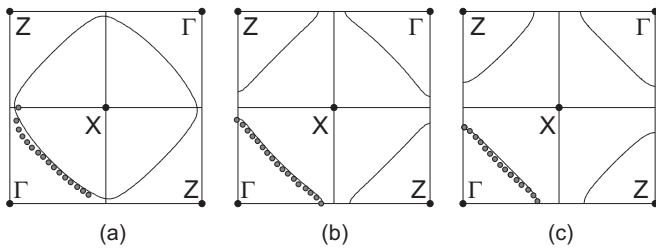


FIG. 4: Comparison of the doping evolution of the measured Fermi surface of $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ from Ref. 48,49 for optimal doping ($x = 0.15$) to overdoping ($x = 0.22$; $x = 0.30$) with the corresponding calculated Fermi surface within the rigid-band approximation of the M31BM, respectively. The dots indicate the experimental results.

photoemission spectroscopy (ARPES) over the doping range considered. Such an agreement obtained within the M31BM and the consistence with the Λ_1 phonons speaks in favour of this model with renormalized c -axis hopping parameters leading to an enhanced anisotropy. Moreover, it demonstrates the importance of an experimental verification of a computed electronic bandstructure. Clearly visible in the experiments and the calculations is the change of the FS topology and of the related nesting structures upon doping when passing from the model OP to model OD1 or OD2, respectively.

Strong nesting of the FS can bring about either a charge density wave or a spin density wave, or possibly both. The tendency toward Fermi surface driven instabilities frequently is announced by maxima of the so called (noninteracting) susceptibility

$$\Pi_0(\mathbf{q}) = -\frac{2}{N} \sum_{n,n'} \frac{f_{n'}(\mathbf{k} + \mathbf{q}) - f_n(\mathbf{k})}{E_{n'}(\mathbf{k} + \mathbf{q}) - E_n(\mathbf{k})}, \quad (26)$$

which can be obtained from Eq. (18) by equating all the expansion coefficients $C_{kn}(\mathbf{k})$ to one. It is obvious from this form that possible FS-nesting at certain \mathbf{q} vectors is reflected as a maximum of this function.

From inspection of the FS in Fig. 4 we can expect that approximate nesting vectors around the X -point of the BZ will lead to corresponding maxima in $\Pi_0(\mathbf{q})$. For the optimally case (model OP) we find a maximum at the X -point and in the overdoped state (model OD1 and OD2) the maximum is progressively shifted toward the Γ point. The position of the maximum for model OP is significant because it coincides with the ordering wave vector in the antiferromagnetic state. For the overdoped state, the ordering wave vector (magnetic scattering peak) is predicted from our calculations for Π_0 to move inward from the X -point. Quite general, in the doped cuprates the spin fluctuations (SF's) are antiferromagnetic in origin and arise from both, nesting properties of the FS (spin-density-wave type SF's) and from correlation driven nearest-neighbour antiferromagnetic superexchange because of the proximity of a long-range ordered antiferromagnetic state in the undoped parent

compounds.

In Fig. 5 we display for the models OP, OD1 and OD2 FS maps for different k_z values ($k_z = 0, k_z = \frac{2\pi}{c}$) as a function of $\mathbf{k} = (k_x, k_y)$. This allows for example to map the k_x -, k_y -dependence of the c -axis dispersion. The calculations demonstrate that the effect of the k_z dispersion vanishes along the nodal direction and that it increases as one moves towards the antinodal region. The three-dimensionality associated with the k_z -dispersion, usually neglected in discussing the physics of the cuprates, has been shown to play a key role in shaping the ARPES spectra^{36,50}, because the residual k_z -dispersion of bands in a quasi-two-dimensional material will induce an irreducible linewidth in ARPES peaks. On the other hand detecting such k_z related linewidth in the ARPES spectra as found for LaCuO ³⁶ establishes the existence of coherent c -axis transport.

Finally to get also a more global impression of the magnitude of the enhanced anisotropy in the M31BM as compared with a typical DFT-LDA based first principles result we compare some FS parameters being important for transport properties like the Drude plasma energy tensor and the Fermi velocity tensor. The Drude tensor is defined as

$$\hbar\Omega_{p,ij} = \left(\frac{8\pi}{NV_z} \sum_{\mathbf{k}n} \delta(E_n(\mathbf{k}) - E_F) v_{\mathbf{k}n,i} v_{\mathbf{k}n,j} \right)^{\frac{1}{2}}, \quad (27)$$

and the Fermi velocity tensor is given by

$$\langle v_{F,ij}^2 \rangle^{\frac{1}{2}} = \left(\frac{2}{N} \sum_{\mathbf{k}n} \Theta(E_n(\mathbf{k}) - E_F) v_{\mathbf{k}n,i} v_{\mathbf{k}n,j} \right)^{\frac{1}{2}}, \quad (28)$$

with

$$\mathbf{v}_{\mathbf{k}n} = \frac{1}{\hbar} \frac{\partial E_n(\mathbf{k})}{\partial \mathbf{k}}. \quad (29)$$

The outcome from our model OP for the optimally doped state is compared in Table I with the corresponding results calculated for optimally doped LaCuO within LAPW^{51,52}. From the table we extract an enhancement of the anisotropy ratio $A_\Omega = \Omega_{p,xx}/\Omega_{p,zz}$ and of $A_{v_F} = (v_{F,xx}^2)^{1/2}/(v_{F,zz}^2)^{1/2}$ by about a factor 5 in the M31BM as compared to the LAPW calculation.

B. Nonadiabation phonon calculations and phonon-plasmon coupling

We prelude this section with a discussion of the anomalous behaviour of the apex-oxygen bond-stretching mode in LaCuO at the Z -point of the BZ (O_z^Z) polarized perpendicular to the CuO plane, see Fig. 6. From the experimental side, it took quite some time until this mode could be assessed³² because of a massive line-broadening of about 4 THz in the metallic phase at optimal doping and its very large softening of about 5.5 THz when passing from the insulating to the metallic state^{31,32}. The

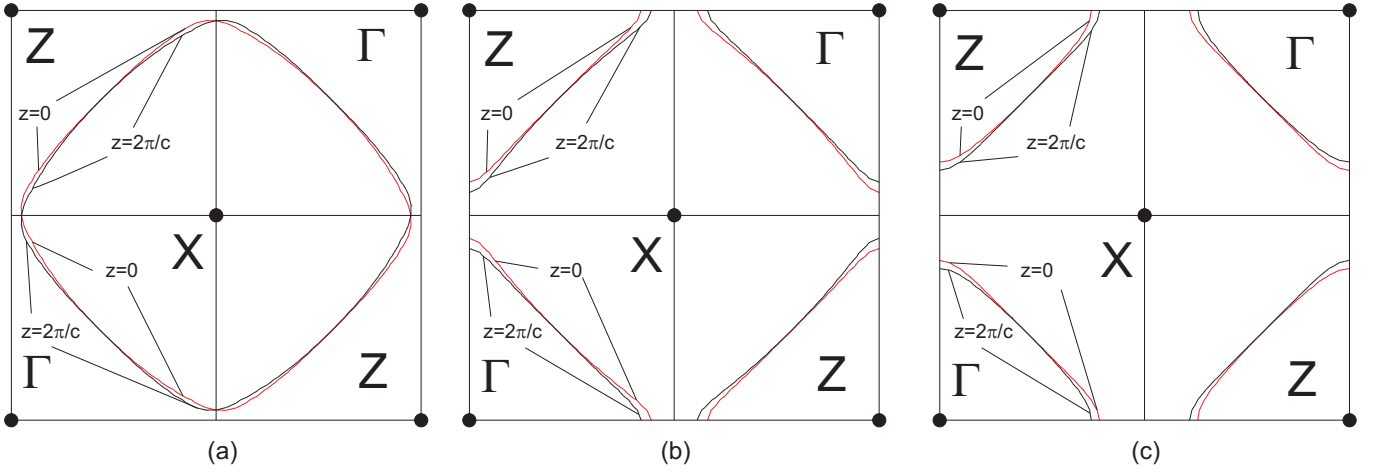


FIG. 5: Fermi surface maps for different k_z values ($k_z \equiv z = 0$ and $k_z \equiv z = \frac{2\pi}{c}$) as a function of $\mathbf{k}_{||} = (k_x, k_y)$ for different doping levels in rigid-band approximation of the M31BM. Model OP ($x = 0.156$) (a), model OD1 ($x = 0.224$) (b) and model OD2 ($x = 0.297$) (c).

	$\hbar\Omega_{p,xx}$	$\hbar\Omega_{p,zz}$	$\langle v_{F,xx}^2 \rangle^{1/2}$	$\langle v_{F,zz}^2 \rangle^{1/2}$	A_Ω	A_{v_F}
LAPW	701.21	132.99	2.20	0.41	5.27	5.37
OP	648.60	25.25	2.99	0.11	25.69	27.97

Table I: Comparison of the Fermi surface parameters (Drude plasma energy tensor, Fermi velocity) and the anisotropy ratios $A_\Omega = \Omega_{p,xx}/\Omega_{p,zz}$; $A_{v_F} = \langle v_{F,xx}^2 \rangle^{1/2}/\langle v_{F,zz}^2 \rangle^{1/2}$ for optimally doped LaCuO between a calculation in LAPW^{51,52} and the M31BM (model OP), respectively. $\Omega_{p,ij}$ is given in units of THz and the Fermi velocity $\langle v_{F,ij}^2 \rangle^{1/2}$ in units of 10^7 cm/s.

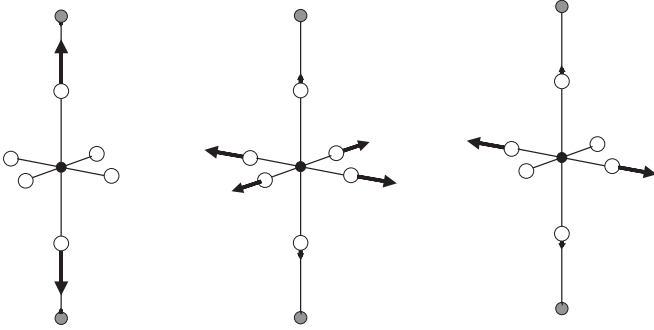


FIG. 6: Displacement patterns of the high-frequency oxygen bond-stretching modes (OBSM) of LaCuO. Left: apex-oxygen breathing mode (O_z^Z), middle: planar breathing mode (O_B^X), right: half-breathing mode ($\Delta_{1/2}$).

strong softening of this mode across the insulator-metal transition was predicted prior to the experimental observation, see Refs.^{28,39,42}.

Optical modes like O_z^Z are of special importance for the cuprates because of their strong *nonlocal* coupling to the electrons due to the long-ranged Coulomb interaction poorly screened along the c -axis in these compounds, even in the metallic state. As a consequence nonlocal, nonadiabatic polar electron-phonon coupling effects

become substantial. These high-frequency c -axis polarized optical phonons are basically unscreened. Quite recently such a long-ranged polar electron-phonon interaction (EPI) has been identified as very essential also for pairing in the cuprate superconductors^{53,54}.

Displacements of the ions in the ionic layers of the cuprates, in particular those of the O_z^Z mode, bring about large changes of the potential felt by the electrons (holes) in the CuO plane which on her part are responsible for the superconductivity and the unusual normal state effects. So, the c -axis polarized optical phonons become important for the renormalization of the electrons in the CuO plane, besides the renormalization due to strong correlation effects. Thus, both, the short-ranged and the long-ranged part of the Coulomb interaction is significant for the physics in the HTSCs.

The manifestation of the coupling between electrons and polar c -axis phonons in the self-energy of the nodal quasiparticles in Bi2201 has been pointed out recently in Refs. 17,55. Note, that the long-ranged Coulomb coupling perpendicular to the CuO plane is very special for the HTSC's and would not be possible in a conventional three-dimensional metal or superconductor because of local screening by a high-density electron gas. As subsequently discussed this long-ranged polar interaction does not only probe the in-plane electron properties but also leads to a strong coupling between electrons and c -axis polarized phonon modes.

In the insulating parent compound O_z^Z is found at about 17 THz while in the optimally doped material the experimental value is estimated at about 11.5 THz³¹. However, the large linewidth of about 3-4 THz^{31,32} and the presence of another Z -point mode at about 14 THz makes it difficult to pin down the exact position of O_z^Z in the metallic phase.

In the following we will demonstrate within the M31BM that due to the nonlocal, nonadiabatic charge

response two coupled phonon-plasmon modes of O_z^Z -type arise at 9.09 THz and 13.86 THz, respectively, and that we can attribute to O_z^Z an *adiabatic* frequency of 12.96 THz. Before discussing these results in more detail we present in retrospect a short explanation of the large softening of O_z^Z during the insulator-metal transition, for details see e.g. Ref. 28 and earlier references therein.

In the insulating state O_z^Z can be shown to induce only an *intralayer* charge transfer between the copper and oxygen orbitals such that local charge neutrality of the cell is maintained under a perturbation due to O_z^Z . In this way the charge rearrangement leading to screening is considerably restricted and thus we find a high calculated frequency of O_z^Z in the insulating state in agreement with experiment. Contrarily, in the metallic state no such restriction is present and O_z^Z generates CF's at Cu and O_{xy} that have the same sign in the whole CuO layer. This ultimately leads to an (instantaneous) interlayer charge transfer in adiabatic approximation which provides an effective screening mechanism for the long-ranged Coulomb interaction and produces the anomalous softening of O_z^Z during the insulator-metal transition in the calculations consistent with the measurements. Eventually, the instantaneous interlayer charge transfer in the adiabatic approximation is replaced by a collective dynamic charge transfer within the phonon-plasmon scenario in a small region around the c -axis, including of course the Z -point. Such a scenario will be investigated next within the M31BM.

In Fig. 7 we present our results of the dispersion of the coupled phonon-plasmon modes in the small region around the c -axis characterized by a nonadiabatic charge response. Specifically, we have calculated the coupled mode dispersion of the Δ'_1 modes from the Z -point along Δ' to the $Z' = (\varepsilon \frac{2\pi}{a}, 0, \frac{2\pi}{c})$ point with $\varepsilon = 0.02$, see Fig. 8. The mixed mode dispersion in Fig. 7(a) is for the optimally doped case of LaCuO (model OP), Fig. 7(b) and 7(c) show the results for the overdoped material, i.e. for model OD1 and OD2, respectively. The strongly coupling O_z^Z modes are indicated as full dots. The broken line is the dispersion of the free-plasmon branch, calculated from Eq. (23) and the dotted line is the borderline for damping due to electron-hole decay, i.e. $\max_{\mathbf{k} \in \text{BZ}} (E_n(\mathbf{k}) - E_n(\mathbf{k} + \mathbf{q}))$ for the band crossing the Fermi level. We observe a slight increase of the frequency of the free-plasmon branch upon doping which indicates a small reduction of the anisotropy.

The phonon-plasmon scenario is exemplified for optimally doped LaCuO in Fig. 7(a) corresponding to the model OP for the proper polarization part. From inspection of the figure we can learn two important facts. Firstly, the range of the region in \mathbf{q} -space with a nonadiabatic charge response can be estimated and, secondly, the massive line broadening of the O_z^Z mode can be understood. The full dots at the Z -point are the phonon-like and plasmon-like $O_z^Z(\text{na})$ mode at 9.09 THz and 13.86 THz, respectively. We denote the lower mode as phonon-like because this mode connects to the adiabatic

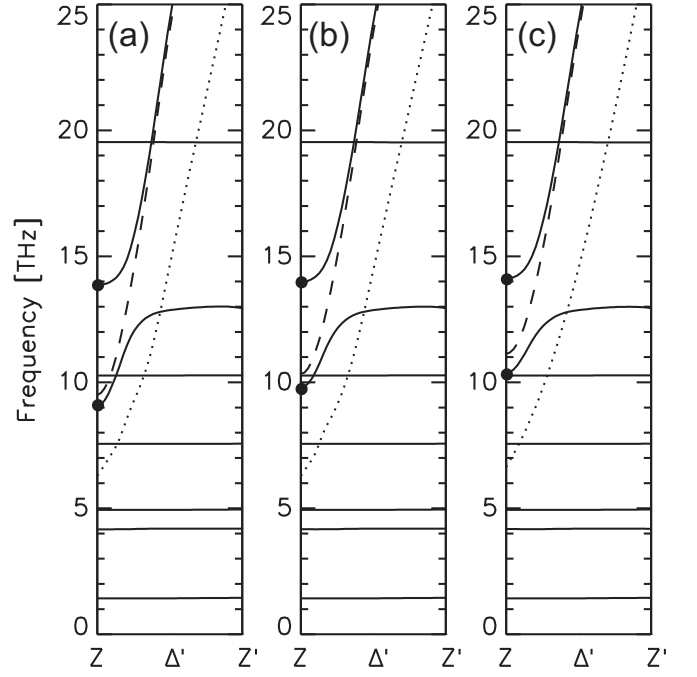


FIG. 7: Calculated coupled phonon-plasmon dispersion of the Δ'_1 , ($O_z^{Z'}$) modes (—) from the Z -point ($\varepsilon = 0$) along $\Delta' = (\varepsilon \frac{2\pi}{a}, 0, \frac{2\pi}{c})$ to $\varepsilon = 0.02$ (Z') for the models OP (a), OD1 (b) and OD2 (c), respectively. The coupling O_z^Z modes at Z are represented by \bullet ; - - : free plasmon branch; \cdots borderline for damping due to electron-hole decay.

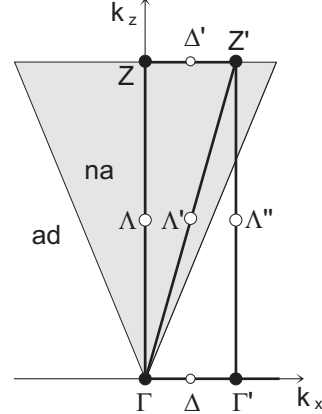


FIG. 8: Schematic representation of the nonadiabatic region in the (k_x, k_z) plane with the directions $\Delta' = (\varepsilon \frac{2\pi}{a}, 0, \frac{2\pi}{c})$, $\Lambda' = \zeta (\varepsilon \frac{2\pi}{a}, 0, \frac{2\pi}{c})$ and $\Lambda'' = (\varepsilon \frac{2\pi}{a}, 0, \zeta \frac{2\pi}{c})$. $\zeta \in [0, 1]$. na: nonadiabatic region; ad: adiabatic region.

$O_z^{Z'}(\text{ad})$ phonon in the adiabatic region. $O_z^{Z'}(\text{ad})$ has virtually the same frequency as the $O_z^Z(\text{ad})$ mode calculated in adiabatic approximation, i.e. $O_z^Z(\text{ad}) = 12.96$ THz. From the figure we can extract that the region of nonadiabatic charge response, characterized by the steep phonon-like branch is very small. It can be estimated at about $\varepsilon \approx 0.01$.

The plasmon-like branch starts at the higher $O_z^Z(\text{na})$ mode, rapidly leaves the frequency range of the phonon spectrum within the nonadiabatic region and becomes more and more plasmon-like as it approaches the free-plasmon dispersion.

The experimentally measured massive line-broadening

for O_z^Z of about 3-4 THz^{31,32} in the optimally doped probe can be understood from the calculated phonon-plasmon scenario displayed in Fig. 7(a). Experimentally, there is a limited wavevector resolution in the transverse direction perpendicular to the c -axis which is on average $\varepsilon \approx 0.03$ (Ref. 56). Thus, the relevant frequency range sampled by the neutron scattering experiment is over the steep branch and is between the two nonadiabatic $O_z^Z(\text{na})$ modes leading to an estimated broadening of about 3.9 THz. Because the region with a metallic, adiabatic charge response outweighs by a factor of three the nonadiabatic region we can attribute to the broad O_z^Z mode an adiabatic frequency of about 12.9 THz.

The calculated character of the coupled modes along Δ' can be obtained from Table II where the displacement amplitudes for the phonon-like and plasmon-like mode is given in terms of relative displacements of the apex oxygen O_z and the La ion along the c -axis. At the Z -point O_z and La are vibrating in phase for the phonon-like mode and out of phase for the plasmon-like mode. In case of the phonon-like mode the amplitude of O_z^Z remains more or less constant while the amplitude for La decreases with increasing ε and finally runs out of phase with O_z in the adiabatic region. In the plasmon-like O_z^Z mode the O_z and La ions always vibrate out of phase. Increasing ε the amplitude of O_z^Z decreases while the magnitude of the amplitude of La increases. The different phase relation of the two modes is reflected in the corresponding induced charge redistributions as discussed below.

The size $\varepsilon \approx 0.01$ of the nonadiabatic region predicted by our calculation within model OP is too small for an experimental observation of the dispersion of the coupled phonon-plasmon modes shown in Fig. 7. Only sampling the nonadiabatic region on average as mentioned above seems possible at present.

Inspection of Fig. 7(b) and Fig. 7(c) for the overdoped states leads to about the same size of the nonadiabatic sector as in the optimally doped case and to the same adiabatic frequency for $O_z^{Z'}(\text{ad})$ and $O_z^Z(\text{ad})$, respectively. However, the broadening of O_z^Z is predicted by the calculation to decrease as compared to the optimally doped case. A decrease of the linewidth of O_z^Z with practically no change of the frequency has also been found quite recently for overdoped LaCuO in the experiments⁵⁷. Thus, our prediction within the rigid-band approximation is conform with the measurements. From Fig. 7(b) and Fig. 7(c) the width of O_z^Z is 3.23 THz for $x \approx 0.22$ and 2.66 THz for $x \approx 0.3$, respectively. Last but not least we can read off from Fig. 7 a doping dependence of the phonon-like O_z^Z mode in which the mode energy decreases with reduced doping (OD2: 10.31 THz; OD1: 9.74 THz; OP: 9.09 THz).

Now we comment on the present interpretation of the measured dispersion of the steep Λ_1 -branch displayed in Fig. 2. We just have shown through our calculation of the phonon-plasmon dispersion along the Δ' direction that the nonadiabatic region around the c -axis is so small

that it cannot be resolved by the INS experiments and only an average of the dispersion can be measured. So, we investigate in detail the coupled mode dispersion along the two directions $\Lambda'' = (\varepsilon \frac{2\pi}{a}, 0, \zeta \frac{2\pi}{c})$ in Fig. 9 and $\Lambda' = (\varepsilon \frac{2\pi}{a}, 0, \frac{2\pi}{c})$ in Fig. 10 nearby the c -axis in order to study the transition from the region of nonadiabatic to adiabatic charge response together with the change of the mode behavior during this transition.

The frequency of the plasmon near Γ' in Fig. 9 is always very high and far outside the range of the phonon spectrum. Consequently the phonon dynamics is adiabatic in this \mathbf{q} -space region. The phonon-like nonadiabatic $O_z^{Z'}$ mode (lower full dot) converges to its adiabatic value (open circle) at about $\varepsilon = 0.01$, while the plasmon-like $O_z^{Z'}$ mode (upper full dot) rapidly leaves the range of the spectrum. Simultaneously the dynamics becomes adiabatic. Compare with the calculation of the phonon dispersion in the right most panel where the static ($\omega = 0$) approximation for $\Pi_{\kappa\kappa'}$ has been used. Likewise as in the investigations along the Δ' direction the nonadiabatic region can be characterized by $\varepsilon \approx 0.01$. and the present resolution limit for INS does not allow to resolve the mixed-phonon plasmon dispersion. Only the result of averaging Λ'' between Γ' and Z' can be detected. In such a procedure the small nonadiabatic part close to the c -axis will be outweighed in the measurement by a significant larger part where the dispersion is nearly adiabatic. In particular we extract the development of the dispersion which leads to the steep Λ_1 branch along Λ found in our calculations in the adiabatic limit. From this discussion it becomes clear how the measured data points in Fig. 2 should be interpreted, namely as an average over the coupled mode dispersion with a dominant adiabatic contribution.

Concerning the steep Λ_1 branch the same conclusion can be drawn from the inspection of the calculated results along the Λ' direction displayed in Fig. 10. Again the phonon-like $O_z^{Z'}$ mode matches the adiabatic result at about $\varepsilon \approx 0.01$ and the dispersion with larger ε values is virtually adiabatic. At the Γ -point two coupling longitudinal A_{2u} modes (full dots) occur which we denote as ferroelectric modes (FM) because of their intrinsic displacement patterns where the oxygen anions are vibrating coherently against the cations in the lattice. As a consequence the electric dipole moments generated by the motion add constructively to a large value. So, we can expect a large oscillator strength and as seen in the experiments¹⁴ the FM dominates the infrared response for polarization along the c -axis not only in the insulating state of LaCuO but also in the well doped metallic state. As already mentioned, such an optical activity in the *metallic* phase cannot be explained using the adiabatic approximation for the calculation of the phonon dispersion as is usually done by applying static DFT for the metal, because there will be no LO-TO splitting (A_{2u} splitting in the present case) being a measure of the oscillator strength in such a calculation.

On the other hand, the observed infrared response in

ϵ	Z(ad)	Z(na)	0.0001	0.001	0.002	0.003	0.004	0.007	0.01	0.02
ω	12.96	9.09	9.08	9.27	9.84	10.59	11.41	12.61	12.86	12.94
$O_z(z)$	0.69	0.66	0.66	0.67	0.69	0.70	0.71	0.69	0.69	0.69
$La(z)$	-0.15	0.25	0.25	0.22	0.14	0.05	-0.04	-0.13	-0.15	-0.15
ω	12.96	13.86	13.86	13.90	14.02	14.28	14.81	18.69	24.38	—
$O_z(z)$	0.69	0.68	0.68	0.68	0.68	0.67	0.67	0.61	0.59	—
$La(z)$	-0.15	-0.20	-0.20	-0.20	-0.21	-0.22	-0.24	-0.33	-0.38	—

Table II: Calculated displacement amplitudes of the phonon-like and plasmon-like $O_z^{Z'}$ modes in model OP along the $\Delta' = (\epsilon \frac{2\pi}{a}, 0, \frac{2\pi}{c})$ direction between $\epsilon = 0$ (Z-point) and $\epsilon = 0.02$. The frequency is given in units of THz. The left most data set is the result for O_z^Z in the adiabatic limit. The plasmon-like mode for $\epsilon = 0.02$ has not been calculated because of its frequency is far out of the relevant frequency range, see Fig. 7(a).

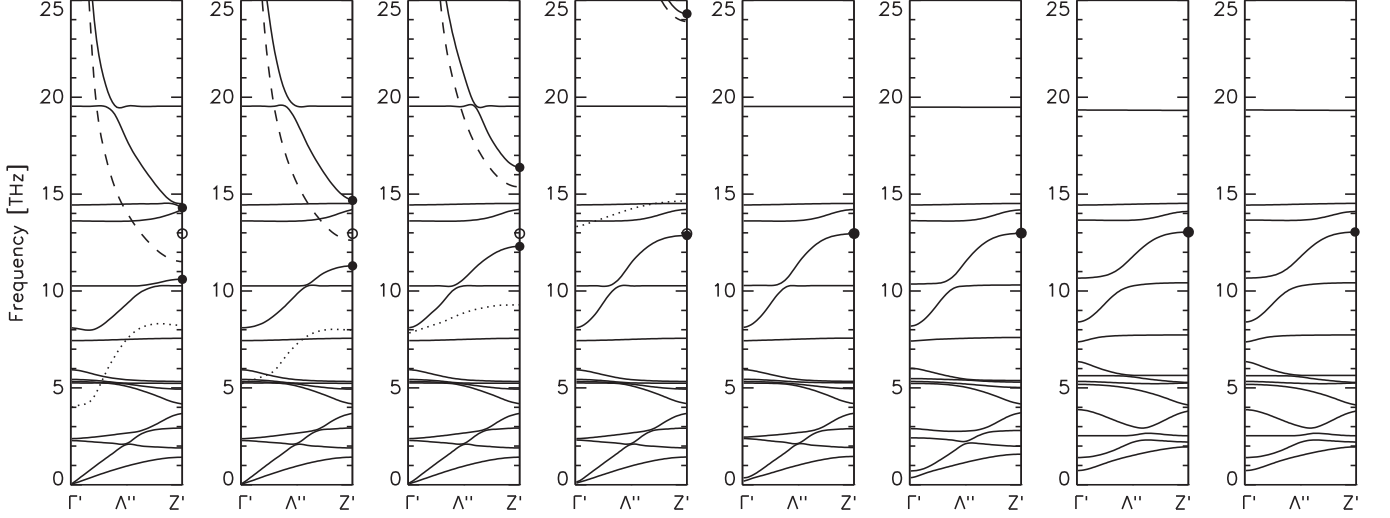


FIG. 9: Nonadiabatic coupled phonon-plasmon dispersion as calculated with model OP for the proper polarization part, representing the optimally doped state of LaCuO from $\Gamma' = (\epsilon \frac{2\pi}{a}, 0, 0)$ to Z' along the $\Lambda'' = (\epsilon \frac{2\pi}{a}, 0, \zeta \frac{2\pi}{c})$ direction. In the different panels of the figure from left to right the following values of ϵ have been used: $\epsilon = 0.003, 0.004, 0.006, 0.01, 0.025, 0.05, 0.100$. The right most panel shows the results of a calculation in adiabatic approximation for $\epsilon = 0.100$. Only the Λ'' branches coupling to the charge fluctuations are shown. —: free-plasmon branch; \cdots : borderline for damping. Dots at Z' : \bullet $O_z^{Z'}(\text{na})$; \circ $O_z^{Z'}(\text{ad})$. na: nonadiabatic; ad: adiabatic.

the metallic phase can be understood from our nonadiabatic results in Fig. 10. Here we find a very large A_{2u} splitting for the longitudinal plasmon-like FM (upper full dot at Γ) at 18.41 THz and the corresponding transverse FM (open circle at Γ) with a calculated frequency of 8.1 THz and 7.4 THz in the experiments⁴³, respectively. Simultaneously, there is a smaller splitting between the latter mode and the longitudinal phonon-like FM (lower full dot at Γ) at 5.80 THz. With increasing ϵ , i.e. when approaching the region of adiabatic charge response, the LO-TO splitting is closed from *below* and the plasmon-like branch with the plasmon-like FM at Γ and the plasmon-like $O_z^{Z'}$ mode at Z' rapidly disappears out of the phonon spectrum. Moreover, the phonon dispersion becomes adiabatic and the signature of the steep Λ_1 branch appears in the Λ' direction.

Summarizing, it should be emphasized that the presence of the large nonlocal long-ranged polar nonadiabatic

electron-phonon coupling which leads to the phonon-plasmon scenario is also reflected in the measured infrared response.

In Fig. 11 we show the calculated nonadiabatic phonon-plasmon dispersion of the Λ_1 modes along the $\Lambda \sim (0, 0, 1)$ direction. The full dots at Γ and Z represent the coupled phonon-like (lower dot) and plasmon-like A_{2u} FM (upper dot) and the phonon-like (lower dot) and plasmon-like (upper dot) O_z^Z mode, respectively. The calculated longitudinal plasmon-like A_{2u} FM at 18.41 THz is not far away from the experimental result for the longitudinal A_{2u} FM of LaCuO in the insulating state obtained with INS at about 17.5 THz. Of course the large splitting between this mode and the corresponding TO mode (open dot) accounts for the dominance of the FM in the infrared response also in the insulating state^{14,43}. It should be remarked that in order to obtain the calculated dispersion curves the effect of the DF's is

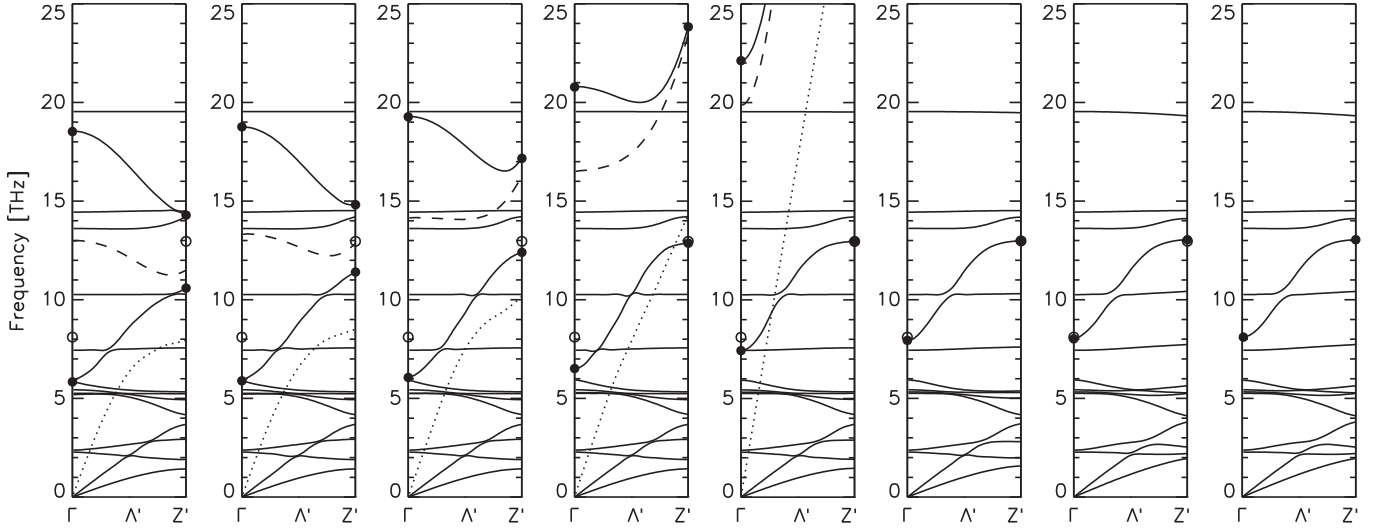


FIG. 10: Nonadiabatic coupled phonon-plasmon dispersion as calculated with model OP for the optimally doped state of LaCuO from Γ to $Z' = (\varepsilon \frac{2\pi}{a}, 0, \frac{2\pi}{c})$ along the $\Lambda' = \zeta (\varepsilon \frac{2\pi}{a}, 0, \frac{2\pi}{c})$ direction. In the different panels from left to right the following values for ε have been used: $\varepsilon = 0.003, 0.004, 0.006, 0.010, 0.025, 0.050, 0.100$. The right most panel shows the result of a calculation within adiabatic approximation for $\varepsilon = 0.100$. Only the Λ' branches (—) coupling to the charge fluctuations are shown. —: free plasmon branch; ···: borderline for damping. Dots at Z' : $\bullet O_z^{Z'}(\text{na})$, $\circ O_z^{Z'}(\text{ad})$. Dots at Γ : $\bullet A_{2u}^{\text{LO}}(\text{ferro, na})$; $\circ A_{2u}^{\text{TO}}(\text{ferro, ad})$. na: nonadiabatic; ad: adiabatic.

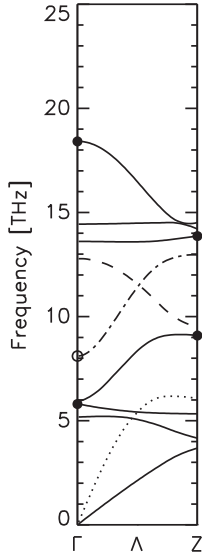


FIG. 11: Calculated nonadiabatic coupled phonon-plasmon dispersion of the Λ_1 modes of model OP for the optimally doped state of LaCuO along the $\Lambda \sim (0, 0, 1)$ direction. The coupling modes at Γ ($A_{2u}^{\text{LO}}(\text{ferro, na})$) and at Z (O_z^Z) are shown as black dots (\bullet). The open dot (\circ) at Γ indicates $A_{2u}^{\text{TO}}(\text{ferro, ad})$. —: Λ_1 modes; —: free plasmon branch; ···: borderline for damping; - · -: steep Λ_1 branch in the adiabatic approximation.

important. The latter reduce the frequency of the plasmon as compared with the calculation which takes only CF's into account. This effect is exemplified in Table III for the free-plasmon and the plasmon-like modes at Γ and Z .

From our preceding discussion of the coupled mode scenario along the Λ' and Λ'' direction it can be concluded that the branch with the phonon-like FM at Γ and the phonon-like $O_z^{Z'}$ mode at Z' converges to the steep Λ_1 branch of Fig. 11 in the Λ direction when approaching the adiabatic limit. Thereby the LO-TO splitting at Γ is closed from below and the phonon-like O_z^Z mode reaches its adiabatic value. The plasmon-like branch connect-

	Γ	Z	Γ	Z
CF	14.09	9.83	20.45	16.99
CFD	12.78	9.51	18.41	13.86

Table III: Frequency in THz of the free c -axis plasmon at the Γ and Z -point of the Brillouine zone (leftmost columns) as calculated with model OP for the optimally doped state. The notation CF means that only charge fluctuations are taken into account; CFD represents a calculation including additionally anisotropic dipole fluctuations. The rightmost columns gives the corresponding results for the plasmon-like modes. Compare with Fig. 11.

ing the corresponding plasmon-like modes at Γ and Z' rapidly leaves the phonon-spectrum as can be seen in Fig. 10.

A comment is appropriate regarding the relation between electronic correlations in the CuO plane beyond LDA and the possible size of the nonadiabatic region which points to an interplay of nonadiabatic effects around the c -axis due to the long-ranged Coulomb interaction and strong short-ranged Coulomb repulsion responsible for correlation. The latter leads in tendency to a reduction of the bandwidth and the Fermi velocity of the quasi-particle (QP) excitations in the (k_x, k_y) -plane as compared with DFT-LDA-like bandstructures⁵⁸. Such a reduction should increase the size of the nonadiabatic region, because the electron dynamics in the CuO plane is slowed down and the free-plasmon frequency is accordingly decreased. In Ref. 59 we have simulated a narrowing of the electronic bandstructure within the 11BM by reducing empirically certain tight binding parameters

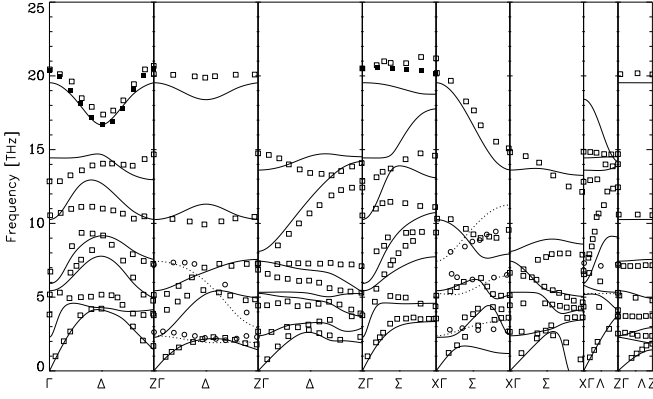


FIG. 12: Calculated nonadiabatic phonon dispersion of optimally doped LaCuO in the main symmetry directions $\Delta \sim (1,0,0)$, $\Sigma \sim (1,1,0)$ and $\Lambda \sim (0,0,1)$ within the M31BM (model OP) for the proper polarization part. The various symbols representing the experimental results from Refs. 31,32 indicate different irreducible representation (ID's). Open symbols are for $\text{La}_{1.9}\text{Sr}_{0.1}\text{CuO}_4$ and solid symbols for optimally doped LaCuO. The arrangement of the panels from left to right according to the different ID's is as follows: $|\Delta_1|\Delta_2(\dots, \circ)$, $\Delta_4(\text{---}, \square)|\Delta_3|\Sigma_1|\Sigma_2(\dots, \circ)$, $\Sigma_4(\text{---}, \square)|\Sigma_3|\Lambda_1(\text{---}, \square)$, $\Lambda_2(\cdot \cdot \cdot \cdot, \circ)|\Lambda_3|$.

and as a result a slight increase of the nonadiabatic region around the c -axis has been found. Thus, it can be expected that characteristic properties of the QP in the CuO plane related to strong correlation effects take influence on the dynamical charge response perpendicular to the CuO layer nearby the c -axis.

Of course the position of the free-plasmon along the c -axis directly depends on the strength of the interlayer coupling which may differ in other cuprates from the situation found for LaCuO in the M31BM. As already mentioned, we have studied in the past parametrically the effect of a varying anisotropy in Refs. 11,12. For a very weak interlayer coupling the resulting nonadiabatic dispersion in the metallic state along the Λ direction virtually cannot be distinguished from the dispersion of the (adiabatic) insulator and the optical phonon modes remain completely unscreened.

As far as the pairing channel via coupled phonon-plasmon modes is concerned it is interesting to remark that the total effective interaction between the electrons via phonon-plasmon exchange is *attractive* in the frequency range between the transverse A_{2u} FM and the longitudinal plasmon-like A_{2u} FM at the Γ point. Thus, the very large splitting of this mode, see Fig. 10 and Fig. 11, is favourable for pairing and the role played in this game by the incomplete dynamical screening of the (bare) polar Coulomb interaction in the nonadiabatic region becomes evident. Note in this context that in recent work⁶ it is found that the phonon-plasmon mechanism contributes constructively and significantly to the superconductivity in the cuprates and that the long-range polar electron-phonon interaction is important for pairing^{53,54}.

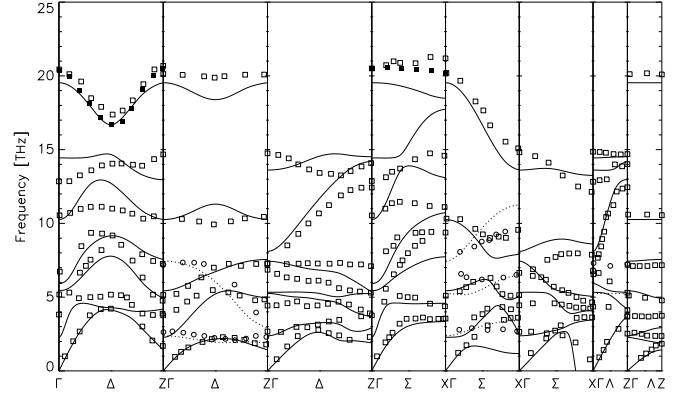


FIG. 13: Same as in Fig. 12, but calculated in the adiabatic approximation.

In Figs. 12, 13 we compare our calculations of the phonon dispersion of LaCuO along the main symmetry directions in the BZ within the M31BM as obtained for the nonadiabatic and adiabatic case of the proper polarization part, respectively. In Fig. 12 the small nonadiabatic region outside the c -axis where the adiabatic approximation grossly fails has been excluded because it cannot be resolved in this figure. Leaving aside the large differences of the calculated dispersion for the Λ_1 modes as already discussed virtually no changes can be observed when passing from the nonadiabatic treatment to the adiabatic calculation. In particular this holds true for the modes propagating in the CuO plane. The reason is the fast electron dynamics in the CuO plane as compared to the dynamics perpendicular to the plane. From all the modes propagating in the CuO layer studied, the O_B^X breathing mode, see Fig. 6, shows the largest deviation, namely a stiffening of 0.284 THz (9.47 cm^{-1}) for optimally doped LaCuO in the nonadiabatic treatment. We attribute this increase of the frequency to a *dynamical* reduced nesting effect with wavevectors around $X = \frac{\pi}{a}(1,1,0)$. Such an interpretation is consistent with the calculated FS in Fig. 4(a) and the maximum found at the X -point for the reduced electronic polarizability function $\Pi_0(\mathbf{q})$ from Eq. (26). The change of the FS topology upon doping from hole-like to electron-like and the related change of the nesting structures in Fig. 4 leads in the calculations to a reduction of the nonadiabatic stiffening of O_B^X (model OD1: 0.193 THz (6.44 cm^{-1}), model OD2: 0.059 THz (1.97 cm^{-1})).

A possible effect of the nonadiabatic charge response on the Raman modes in LaCuO could be quite interesting because very recently giant nonadiabatic effects in layered metals such as graphite intercalated compounds, which are three-dimensional metals with a considerable anisotropy along one direction, have been predicted in Ref. 60. Contrarily, in our calculations for LaCuO practically no nonadiabatic effects have been found for the Raman modes.

Finally, a comment on the calculated unstable mode in Figs. 12, 13 at the X -point should be made. This mode is

the so called tilt mode. Freezing of this distortion points correctly to the experimentally observed structural phase transition from the high-temperature tetragonal (HTT) to the low-temperature orthorhombic (LTO) structure. In earlier calculations for the LTO structure we have found within the ab initio RIM two stable tilt modes with different frequencies in reasonable agreement with the experiment¹². From these calculations it can be concluded that the structural phase transition is essentially driven by the strong long-ranged Coulomb interactions between the ions in the material which again emphasizes the important role of ionic binding for the cuprates.

C. Phonon-plasmon induced charge response and selfconsistent changes of the potential

In previous work^{12,61,62} we have made explicit the charge response by calculating in adiabatic approximation the phonon-induced charge density redistribution for the presumably generic phonon anomalies $\Delta_1/2$ (half-breathing mode) and O_B^X (planar-breathing mode), essentially polarized in the CuO plane and also for the O_z^Z mode²⁸ polarized perpendicular to the plane, see Fig. 6 for the displacement patterns of these oxygen-bond-stretching modes (OBSM). We have shown in Sec. B that $\Delta_1/2$ and O_B^X can be treated in the adiabatic limit, but this is of course not true for O_z^Z as a specific important mode from the nonadiabatic region of charge response.

As suggested in Ref. 63 specific modes (in particular $\Delta_1/2$) with a strong electron-phonon coupling can generate an *overscreening* of the intersite Coulomb interaction due to a phonon-induced charge transfer between Cu and O_{xy} . This strong coupling has been proposed in⁶³ to form a basis for the phonon mechanism of high-temperature superconductivity.

The anomalous softening of the OBSM leading as in case of $\Delta_1/2$ to a decreasing dispersion when starting at the Γ point, see Figs. 12,13, is a result of *overscreening* at shorter wave lengths of the changes of the Coulomb potential generated by the motion of the ions. This has been shown by our calculations to be due to nonlocally excited ionic CF's localized at the Cu and O_{xy} sites generating a dynamic charge ordering in form of localized stripes of alternating sign in the CuO layer. Such induced charge patterns also have been calculated quite recently for *n*-doped NdCuO²⁹. The importance for the phonon anomalies $\Delta_1/2$ and O_B^X of the repulsive on-site Coulomb interaction U_d as a relevant physical parameter of the cuprates deciding about the correlations in these materials has been predicted already in Ref. 39 and investigated in more detail in Refs. 28,61. Briefly, a large U_d suppresses double occupancy of the Cu3d orbital and thus Cu3d CF's which are crucial for the softening of O_B^X and $\Delta_1/2$.

For the $\Delta_1/2$ mode the charge stripes point along the CuO bonds and for O_B^X along the diagonals of the CuO plane. Moreover, in Ref. 12 we have pointed out by

means of the OBSM, how antiferromagnetic correlations may favour the CF's in these modes and vice versa that nonlocal EPI is expected to generate antiferromagnetic spin-fluctuations (SF's) via the phonon-induced CF's between the Cu ions. This interplay between charge- and spin-degrees of freedom should die out in the overdoped state because of the vanishing of the antiferromagnetic correlations.

The induced charge redistribution for O_z^Z calculated in the adiabatic limit has been addressed shortly in Sec. B and will be compared explicitly in Fig. 14 with the corresponding results as obtained in the nonadiabatic approach. So far we can say that the phonon anomalies $\Delta_1/2$ and O_B^X reflect the importance of the short-ranged part of the Coulomb interaction (U_d) while O_z^Z is specifically governed by the long-ranged part.

Before proceeding with the investigation of the charge response in the optimally to overdoped metallic state of LaCuO we will include for completeness a short summary of our approach to phonon dynamics and charge response in the insulating and underdoped state of the cuprates. A detailed discussion can be found in Refs. 12,28,29,61,64. In this work also the strong *doping dependence* of the OBSM phonon anomalies has been studied.

In order to describe these states of the cuprates we have proposed a microscopic modeling which takes into account the enhanced correlation of the Cu3d orbitals in these "phases" via a compressibility-incompressibility transition for the Cu3d state in terms of rigorous sum rules for the proper polarization part $\Pi_{\kappa\kappa'}(\mathbf{q})$ in the longwavelength-limit. An interesting aspect of the partial incompressibility of the Cu3d states is that in case pairing correlations (pre-pairing) would exist above T_C the reduced local particle number fluctuations related to the orbital incompressibility of Cu3d due to the large on-site repulsion would correlate with enhanced phase fluctuations according to the particle number phase uncertainty relation and consequently a collective onset of phase coherence is suppressed. This in turn would stabilize a pre-paired state above T_C .

In Refs. 29,64 also the relation of the superconducting gap and the pseudogap as well as the possibility of Fermi surface transformations observed in the normal state of the cuprates is discussed within the model. Moreover, the modeling has been extended to *n*-doped cuprates and the *electron-hole asymmetry* is simulated via an orbital selective incompressibility-compressibility transition of the O_{2p} orbital in contrast to the *p*-doped case.

We now return to the investigation of the phonon-induced nonadiabatic charge response in optimally doped LaCuO represented by the model OP. The nonlocal, non-rigid part of the charge response related to the nonlocal EPI effects as excited by a phonon with wavevector \mathbf{q} and polarization σ is given by

$$\delta\rho_n(\mathbf{r}, \mathbf{q}\sigma) = \sum_{\mathbf{a}, \kappa} \delta\zeta_{\kappa}^{\mathbf{a}}(\mathbf{q}, \sigma) \rho_{\kappa}(\mathbf{r} - \mathbf{R}_{\kappa}^{\mathbf{a}}). \quad (30)$$

The CF's $\delta\zeta_{\kappa}^{\mathbf{a}}$ are obtained from Eq. (19) and the form-

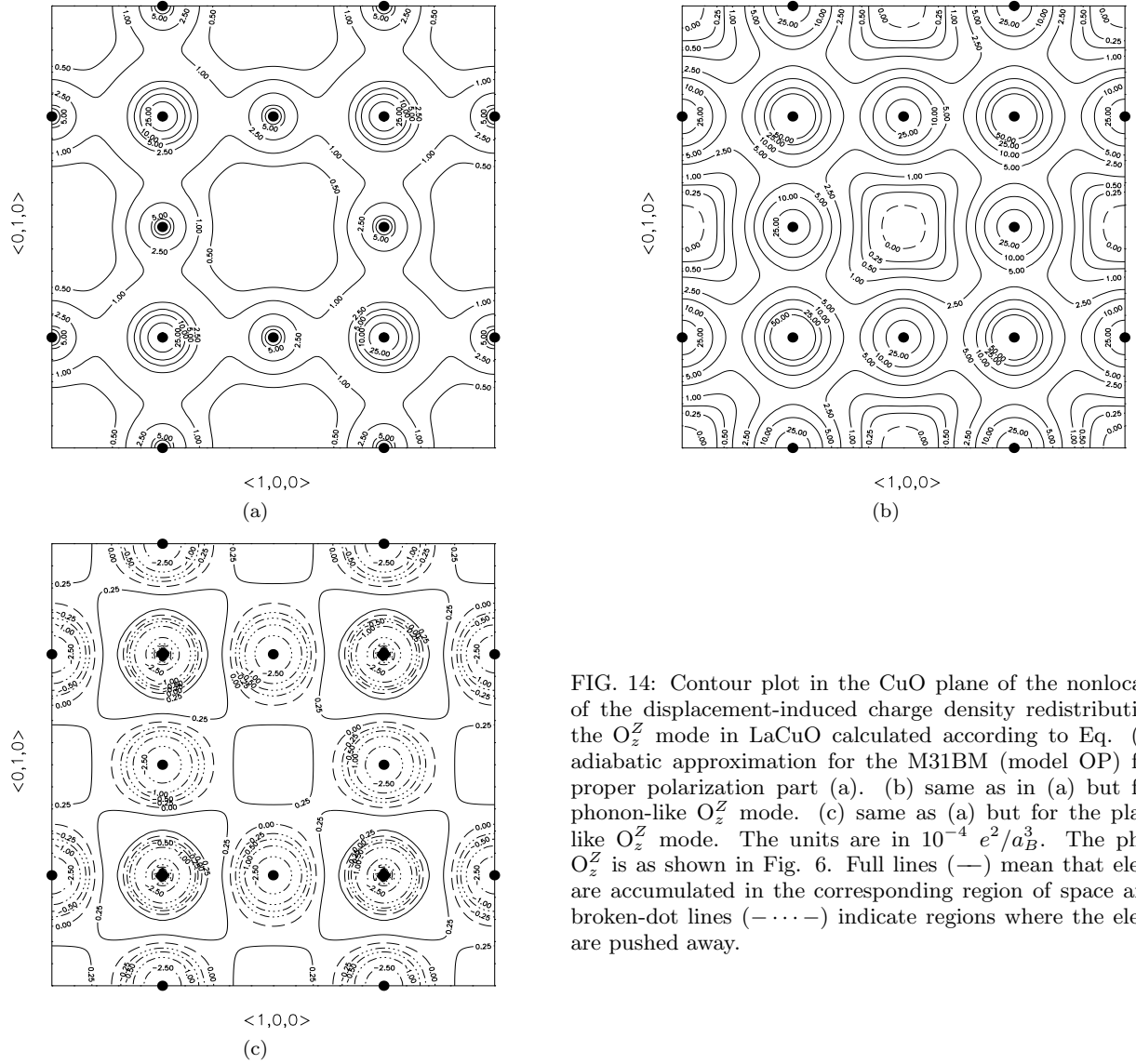


FIG. 14: Contour plot in the CuO plane of the nonlocal part of the displacement-induced charge density redistribution for the O_z^Z mode in LaCuO calculated according to Eq. (30) in adiabatic approximation for the M31BM (model OP) for the proper polarization part (a). (b) same as in (a) but for the phonon-like O_z^Z mode. (c) same as (a) but for the plasmon-like O_z^Z mode. The units are in $10^{-4} e^2/a_B^3$. The phase of O_z^Z is as shown in Fig. 6. Full lines (—) mean that electrons are accumulated in the corresponding region of space and the broken-dot lines (— · — ·) indicate regions where the electrons are pushed away.

factors ρ_K from Eq. (2).

In the contour plots of Fig. 14 the displacement induced rearrangement of the charge density according to Eq. (30) is shown for the O_z^Z mode. The calculation has been performed for the model OP. Fig. 14(a) displays the result for the adiabatic approximation and Fig. 14(b) and Fig. 14(c) pictures the nonadiabatic result for the phonon-like and plasmon-like mode O_z^Z mode, respectively. The phase of the vibration of the ions is as in Fig 6, i.e. the apex oxygens move away from the CuO layer. The full lines ($\delta\rho_n > 0$) indicate that electrons are accumulated in the associated region of space. The broken lines ($\delta\rho_n < 0$) mean that electrons are depleted in this region.

As already mentioned in Sec. B and displayed explicitly in Fig. 14(a) in the adiabatic approximation O_z^Z (12.96 THz) generates CF's at the Cu and O_{xy} ions of the same sign in the whole CuO layer. In the adjacent layers the induced CF's are the same in magnitude but

of opposite sign, compare with Fig. 15(a). Thus, O_z^Z activates an interlayer charge transfer which is instantaneous in adiabatic approximation. Such a nonlocally excited screening process is very effective for screening the changes of the Coulomb interaction generated by the displacement of the apex oxygen ions and explains accordingly the large softening of O_z^Z during the insulator-metal transition.

Fig. 14(b) records our calculated results for the nonadiabatically induced CF's of the phonon-like O_z^Z mode in the CuO plane at 9.09 THz. In the nonadiabatic regime the electrons cannot follow instantaneously the movement of the ions. The now dynamically excited CF's at Cu and O_{xy} have the same sign in the CuO layer. The sign is as in the adiabatic case because the frequency of the free plasmon at Z is (slightly) higher (9.5 THz, see Table III) than the phonon-like O_z^Z mode. The magnitude of the CF's is considerably larger in the nonadiabatic case, see also Table IV.

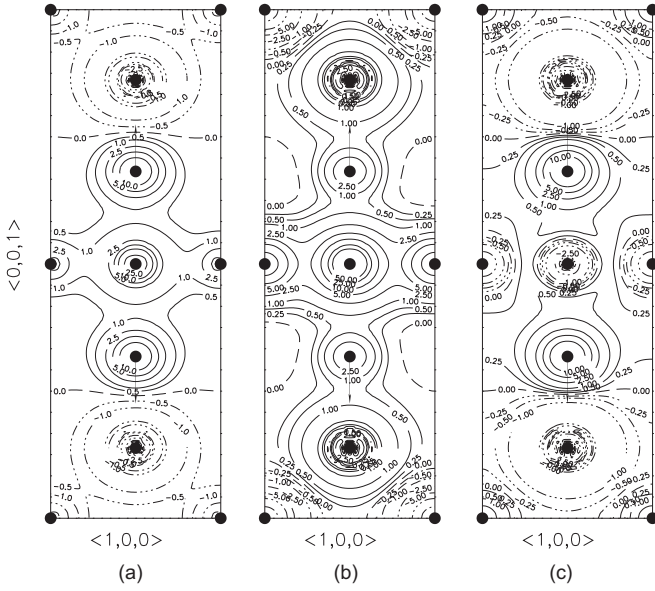


FIG. 15: Contour plot perpendicular to the CuO plane of the nonlocal part of the displacement-induced charge density redistribution for the O_z^Z mode in LaCuO calculated from Eq. (30) in adiabatic approximation for the M31BM (model OP) (a). (b) same as (a) but for the phonon-like O_z^Z mode. (c) same as (a) but for the plasmon-like O_z^Z mode. The units and the meaning of the line types as in Fig. 14. The arrows denote the displacements of the apex oxygen ions.

The interlayer CT is now of dynamic nature because of the dynamical screening of the Coulomb interaction and consequently a coupled collective phonon-like excitation propagating along the c -axis is created leading to the *correlated* charge rearrangement between the layers due to the *coherent* dynamics as shown in the snapshot of Fig. 15(b). Comparing with the adiabatic limit besides the strongly enhanced charge response, which explains the lower frequency, the sign of the CF's at the La ion has changed as compared with the adiabatic calculation. This correlates well with the change of sign of the displacement amplitude for La, compare with Table II. On the other hand, the sign of the CF's at the La for the plasmon-like mode is the same as in the adiabatic case in agreement with the same sign of the displacement of La in both calculations.

The coupled plasmon-like O_z^Z mode at 13.86 THz has a higher frequency than the free plasmon and apparently the charge response runs out of phase, see Fig. 14(c). This results in an *antiscreening* consistent with the higher mode frequency. The sign of the CF's at Cu and O_{xy} is not only opposite to that in the adiabatic and phonon-like case but also smaller in magnitude (Table IV). Fig. 14(c) together with Fig. 15(c) give a snapshot of the collective plasmon-like interplane charge excitations being out of phase with the lattice and also strongly reduced in magnitude as compared with the phonon-like excitation displayed in Fig. 14(b) and Fig. 15(b), respectively.

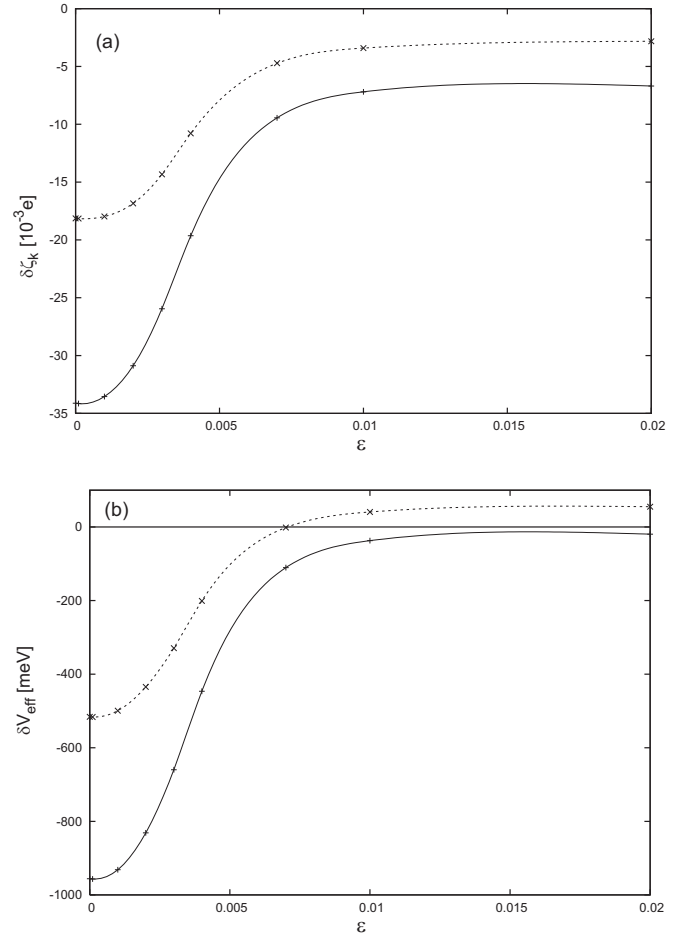


FIG. 16: Calculated displacement-induced charge-fluctuations $\delta\zeta_{\kappa}(\mathbf{q}, \sigma, \omega)$ (Eq. (19)) (a) and corresponding orbital averaged changes of the crystal potential $\delta V_{\kappa}(\mathbf{q}, \sigma, \omega)$ (Eq. (22)) (b) for the phonon-like $O_z^{Z'}$ mode along the Δ' direction ($\varepsilon \frac{2\pi}{a}, 0, \frac{2\pi}{c}$), $\varepsilon \in (0.00, 0.02)$. Cu3d (—) and $O_{xy}2p$ (···) orbital degrees of freedom are considered. The calculations have been performed for model OP.

Concerning c -axis transport in a highly anisotropic metal it is interesting to note that in a simplified model treatment the possibility has been pointed out that a strong coupling between electrons and a bosonic mode propagating and polarized in the c -direction, like O_z^Z , can lead to a metallic to nonmetallic crossover in the c -axis resistivity as the temperature is increased with no corresponding feature in the xy plane properties⁶⁵.

In order to get an impression for the displacement induced changes of the CF's $\delta\zeta_{\kappa}^{\text{a}}(\mathbf{q}, \sigma)$ according to Eq. (19) and the corresponding induced orbital averaged changes of the potential felt by the electrons $\delta V_{\kappa}^{\text{a}}(\mathbf{q}, \sigma)$ from Eq. (22), when passing from the region with a nonadiabatic dynamic charge response to the region with practically static adiabatic charge response, we display in Figs. 16(a,b) our calculated results for the phonon-like $O_z^{Z'}$ mode along the Δ' direction, compare with Fig. 7(a). The calculations have been carried out for the optimally

doped case using model OP and for the most important Cu3d and $O_{xy}2p$ orbitals, respectively. Similar as for the phonon frequencies in Fig. 7(a) $\delta\zeta_\kappa$ and δV_κ converge beyond $\varepsilon \approx 0.01$ to their values in the adiabatic limit. Taking for example the calculated numbers at $\varepsilon = 0.00$ and $\varepsilon = 0.02$, respectively, we recognize that the nonlocally excited CF's and in particular the nonlocally induced orbital averaged changes of the potential being a measure of the long-ranged polar coupling of the electrons and phonons are very strongly enhanced in the nonadiabatic region of phase space around the c -axis.

Additionally, in Table IV we summarize and compare the calculated orbital selective values of $\delta\zeta_\kappa$ and δV_κ for Cu3d and $O_{xy}2p$ of the phonon anomalies $\Delta_1/2$, O_B^X and O_z^Z , respectively. We extract already strong coupling of the adiabatic modes $\Delta_1/2$, O_B^X and of course the strong enhancement for the phonon-like O_z^Z mode. This again points to the fact that besides the short-ranged part of the Coulomb interaction U_d being important for $\Delta_1/2$ and O_B^X , the strong long-ranged, nonlocal polar Coulomb interaction must play an important role in the normal and superconducting state of the doped cuprates.

In context with the very strong coupling of the *doping dependent* phonon-like O_z^Z mode which is at 9.09 THz (37.6 meV) in model OP for the optimally doped state a special remark should be made. This mode involves momentum transfer between antinodal regions of the FS (similar as O_B^X or the resonant magnetic mode), however, with a drastically enhanced coupling strength as compared to O_B^X , see Table IV. Consequently, this mode should contribute to the electron self-energy via electron-phonon coupling in the antinodal region and possibly also to the antinodal pseudogap in the normal state. In the superconducting state even an increased contribution to the self-energy via O_z^Z can be expected because of the large density of states enhancement in these regions due to the opening of a gap with d -wave symmetry⁶⁶. Moreover, below T_C we will have a suppression of the low scattering rate of the electrons due to the opening of a gap and additionally below the finite energy of the phonon-like mode.

This might be helpful to understand the dramatic change in lineshape of the antinodal spectra seen in ARPES, where a sharp quasi-particle peak develops at the lowest binding energies followed by a dip and a broader hump. In the current literature these features are explained by a finite energy collective bosonic mode which mostly is identified as the magnetic resonance mode^{67,68}. On the other hand, the enhanced coherence of the quasi-particles at lower temperature in the superconducting state is simultaneously favourable for the existence of a phonon-plasmon scenario with the plasmon-like mode as another possibility for the collective bosonic mode.

From our calculations damping of the plasmon due to electron-hole decay is less important because only intraband excitations contribute in case of the simple BS near the Fermi level. So, the phonon-like mode should

be an important low-energy bosonic excitation mediating an attractive interaction. Comparing with the isostructural low-temperature superconductor SrRuO these relations can be expected to change considerably because of the complex BS around the Fermi energy where three FS sheets exist^{69,70,71}. In such a situation a strong \mathbf{q} -dependent plasmon damping by low-lying interband transitions can be expected in the collisionless regime and the c -axis plasmon might cease to be a well defined excitation. Note, that in simple metals it has been shown that interband transitions are the dominant processes in the damping of plasmons⁷². Additional damping due to interactions between electrons and with other degrees of freedom enhances the damping still further and broadens the energy levels. This leads to a narrowing of the interband gaps and to the fact that the decay via interband transitions would even be promoted. Altogether, it seems likely that in contrast to LaCuO in the low-temperature superconductor SrRuO the c -axis phonon-plasmon modes are not well defined. On the other hand, the existence of these modes in the cuprates could point to their relevance for high-temperature superconductivity.

In case of cuprate superconductors with more than one CuO layer a qualitatively new possibility may arise as candidate for the finite energy collective bosonic mode. In these compounds additionally to the low-lying plasmon (intraband plasmon) of the single layer compounds discussed so far which results from a weak but non vanishing k_z dispersion of the QP's an interband plasmon with a low energy can exist. The latter emerges at vanishing k_z dispersion from a finite but small intra-layer coupling, e.g. between the two CuO layers in a double layer system. Such a possibility has been investigated qualitatively for YBaCuO within a simplified model approach, see Refs. 12,73 analogously to the model for LaCuO in Refs. 11,12.

In case of YBaCuO a 22BM is used and then the degeneracy of the BS is lifted by introducing parametrically an intra-bilayer coupling. The latter is varied to simulate a corresponding bilayer splitting. According to these calculations a sufficiently weak bilayer coupling generates a nearly dispersionless low-lying phonon-like bond bending branch polarized along the c -axis with a phonon-like $A_g^Z(O_{23} \uparrow\uparrow)$ mode at the Z -point and a longitudinal phonon-like FM ($B_{1u}^\Gamma(O_{23} \uparrow\uparrow)$) at the Γ -point. The notation ($O_{23} \uparrow\uparrow$) means that the O_2 and O_3 oxygen ions in the CuO layer are vibrating in phase. With increasing bilayer-coupling the frequency of the phonon-like $A_g^Z(O_{23} \uparrow\uparrow)$ varies between around 35 meV at weak coupling and 46 meV in the adiabatic limit.

The calculated phonon-induced charge rearrangement of $A_g^Z(O_{23} \uparrow\uparrow)$ in YBaCuO is similar to that of O_z^Z in LaCuO as far as a double layer of YBaCuO is concerned. The CF's have the same sign in the CuO plane but are of opposite sign in the two CuO planes in the double layer. This is energetically favourable because of the attractive Coulomb interaction at short distance between the CF

	$O_z^Z(\text{ad})$		$O_z^Z(\text{na,ph})$		$O_z^Z(\text{na,pl})$		$O_E^X(\text{ad})$		$\Delta_1/2(\text{ad})$		$O_E^X(\text{na})$		$\Delta_1/2(\text{na})$	
	Cu3d	O2p	Cu3d	O2p	Cu3d	O2p	Cu3d	O2p	Cu3d	O2p	Cu3d	O2p	Cu3d	O2p
$\delta\zeta_\kappa$	-6.59	-2.63	-34.12	-18.13	1.32	2.13	-16.45	0	-14.76	-1.44	-16.95	0	-14.76	-1.44
δV_κ	-15.77	58.67	-955.98	-515.98	241.34	207.04	-93.92	0	-73.06	55.14	-87.20	0	-73.13	55.08

Table IV: Magnitudes of the charge fluctuations $\delta\zeta_\kappa$ (Eq. (19)) and orbital averaged changes of the selfconsistent changes of the crystal potential δV_κ (Eq. (22)) of the OBSM as calculated with model OP for the Cu3d and the O2p orbital in the CuO plane. $\delta\zeta_\kappa$ is given in units of 10^{-3} particles and δV_κ in units of meV. na: nonadiabatic; ad: adiabatic; ph: phonon-like; pl: plasmon-like. The phase of the displacements for the OBSM is as shown in Fig. 6. $\delta\zeta_\kappa < 0$ means an accumulation of electrons in the corresponding orbital. $\delta V_\kappa > 0$ indicates that the region around the ion is attractive for electrons.

redistribution of the two layers. So, this CF pattern also explains the strong renormalization of the frequency of this mode as compared with a calculation where the CF's and the related coupling within de bilayer is ignored (61 meV) and DF's only are considered⁷³.

Concerning the infrared response the weak bilayer-coupling only leads to a partial screening of the long-ranged Coulomb interaction and thus to a corresponding fractional reduction of the LO-TO splittings, i.e. the latter are still present consistent with the measured optical *c*-axis spectra in YBaCuO, see e.g. Refs. 74,75. However, the splittings are gradually closed from below by admitting additionally to the intra-bilayer coupling an inter-bilayer coupling in the model leading to a non vanishing k_z dispersion and, finally, to the complete metallic screening of the long-ranged Coulomb interaction in the adiabatic limit.

The $\Delta_1/2$ anomaly involves momentum transfer between nodal regions and could be important to understand the corresponding self-energy corrections, in particular, the nodal kink observed in ARPES experiments. Indeed, in a recent bulk-sensitive low photon energy ARPES study⁷⁶ it has been demonstrated that electron-phonon coupling is responsible for the nodal kink and the "half-breathing" mode $\Delta_1/2$ is the relevant mode.

Another general aspect becomes evident from our calculations namely that the specific features of the solid-state chemistry of the cuprates are essential. The distinctive ionic character of these compounds underlines the relevance of the long-ranged polar Coulomb interaction also in the doped metallic state. On the other hand, the Mott insulating character of the undoped parent compounds being antiferromagnetic insulators emphasizes the importance of the short-ranged part of the interaction, notably by the repulsive on-site interaction U_d . As discussed in this work the impact of both parts of the interaction appears when studying the generic OBSM phonon anomalies.

In the overwhelming number of attempts to describe the physics of the cuprates the short-ranged part U_d is considered exclusively and simplified models with strongly reduced Hamiltonians are used. Such models in the first place may be useful for the discussion of theoretical ideas in general rather than for realistic calculations where the material specifica must be included. Thus, at present it is not at all clear if the two-dimensional

one-band Hubbard model which is most commonly applied for the cuprates contains sufficient information to describe the real materials. In this context a recent combined DFT and dynamical cluster Monte Carlo calculation for the three-band Hubbard model is instructive and may serve as a guideline⁷⁷ in the discussion of a minimal model for the cuprates. This study demonstrates that the corresponding phase diagram of the model and in particular a possible occurrence of superconductivity is quite sensitive to the choice of hopping parameters and the down-folding procedure used. In a recent study⁷⁸ pairing correlations on doped Hubbard models are reanalyzed using a sign-problem-free Gaussian-Basis Monte Carlo method and it is concluded that the simple Hubbard model does not account for high-temperature superconductivity.

The strong short-ranged repulsive U_d by itself does not help with the pair-binding, however, favours *d*-wave symmetry and the usual argument then is that as an implication of a large U_d an antiferromagnetic exchange coupling J arises which leads to an attractive interaction between electrons of opposite spins on neighbouring sites. U_d is an unretarded particle-particle interaction with no low-frequency dynamics and also the interaction related to J is unretarded.

On the other hand, several retarded attractive interactions mediated by virtual bosonic excitations with low-frequency dynamics are discussed in context with pairing in the cuprates. Most common are fluctuations of spin and/or charge degrees of freedom of the electron liquid and (adiabatic) phonons. From our findings in this paper low-frequency coupled phonon-plasmon modes resulting from the strong polar long-ranged interaction in the nonadiabtic region should be added to the list. It still remains an open question which contribution and what kind of cooperation between the different players is most important for pairing and superconductivity in the cuprates. It seems that the high as well as the low energy scale is essential for a synergetic interplay of charge-spin and lattice degrees of freedom which underlies the physics in the cuprates in the normal as well as in the superconducting state.

IV. SUMMARY AND CONCLUSIONS

We have developed a realistic description of the electronic bandstructure of LaCuO which is well suited to calculate the wavevector and frequency dependent proper polarization part of the DRF for the optimally to overdoped state. The latter is used to calculate the adiabatic and nonadiabatic charge response and the coupled mode dynamics in LaCuO.

The large anisotropy along the c -axis of the electronic structure of the cuprates is considerably underestimated in DFT-LDA calculations. So we have modified a LDA-based 31BM to account for the much weaker interlayer coupling in the real material. We have optimized the interlayer coupling by reducing some relevant hopping parameters perpendicular to the CuO layer in such way that significant features of the Λ_1 phonons polarized parallel to this axis are well described. The Λ_1 modes have been chosen because they are most sensitive in respect to the charge response orthogonal to the CuO plane. The new sufficiently anisotropic model (M31BM) leads to a more extended saddle point region, a reduction of the width of the bandstructure, flatter bands in c -direction and a significant narrowing and increase of the important van Hove peak. Simultaneously, we obtain an associated amplification on the average of the proper polarization part. These results demonstrate the importance of a correct representation of the interlayer couplings being relevant for both the electronic properties in the CuO layers and along the c -axis.

Within the M31BM the measured Fermi surfaces for optimally and overdoped LaCuO are well described. Clearly visible in the experiments and the calculations is the change of the FS topology and of the nesting structures upon doping. The latter are shown to be reflected in the (noninteracting) susceptibility. Moreover, relevant Fermi surface parameters for transport like the Drude plasma energy tensor and the Fermi velocity tensor have been calculated and compared with standard LAPW calculations. An enhancement of about a factor of 5 for the anisotropy ratio is found for both types of parameters in the M31BM.

In our calculation of the phonon dispersion of the c -axis polarized Λ_1 modes dramatic changes occur between the adiabatic limit based on a static DRF and the nonadiabatic calculation founded on a dynamic DRF. The former leads to static and the latter to dynamic screening of the Coulomb interaction. On the other hand, virtually no nonadiabatic effects have been detected for the phonons propagating in the main symmetry direction Δ and Σ in the CuO plane. This is due to the fast dynamics of the electrons in the CuO layer as compared with the slow dynamics perpendicular to the plane. Only the planar oxygen breathing mode O_B^X experiences a minor nonadiabatic correction worth mentioning which we attribute to dynamical reduced nesting.

The adiabatic approximation as a basis paradigma to investigate lattice vibrations fails severely in a small sec-

tor of \mathbf{q} -space around the Λ direction. The large nonadiabatic effect found for the Λ_1 modes results from a low-lying c -axis plasmon predicted within the M31BM. This collective mode couples via the strong long-ranged polar Coulomb interaction to the optical phonons of allowed symmetry. In particular we have shown that the large softening of the O_z^Z mode during the insulator-metal transition and its giant linewidth found experimentally can be well understood in the phonon-plasmon scenario.

At the Γ point we identify ferroelectric-like A_{2u} modes with a large LO-TO splitting in form of plasmon-like and phonon-like excitations. As seen in the experiments the ferroelectric-like A_{2u} mode dominates the infrared response for polarization along the c -axis not only in the insulating state of LaCuO but also in the well doped metallic state. Such an optical activity also found in our nonadiabatic calculations for the metallic phase cannot be explained using the adiabatic approximation with static screening for the calculation of the phonon dispersion. This is what has been routinely done in the literature in first principles calculations within static DFT. So, the latter are not consistent with experimental evidence concerning the c -axis response.

From our systematic investigation of the nonadiabatic charge response and phonon dispersion we can identify a small region in \mathbf{q} -space around the Λ direction where the adiabatic approximation breaks down. In this sector of \mathbf{q} -space the strong nonlocal, nonadiabatic polar electron-phonon coupling leads to a mixed phonon-plasmon dispersion. In order to experimentally resolve the calculated dispersion a very high resolution perpendicular to the c -axis would be needed. Presently, inelastic neutron scattering experiments with such a high resolution are out of range and the actual measurements only perform some average orthogonal to the c -axis where the adiabatic charge response dominates by far because of the smallness of the nonadiabatic zone.

We have calculated the induced charge response of the apex oxygen stretching mode O_z^Z in the adiabatic limit and the nonadiabatic phonon-plasmon regime. Additionally we have computed the associated orbital averaged changes of the potential felt by the electrons when passing from the nonadiabatic phonon-plasmon region to the adiabatic regime. In the adiabatic case we have an instantaneous interlayer charge transfer while in the nonadiabatic situation the interlayer charge transfer is dynamic in nature and phonon-like and plasmon-like excitations are excited. Comparing with the adiabatic result we find for the phonon-like O_z^Z mode a strongly enhanced in phase charge response which may be denoted as overscreening with respect to the adiabatic case and accordingly a lower frequency appears while for the plasmon-like mode with a higher frequency the charge response is significantly weaker and runs out of phase, i.e. we observe an antiscreening effect.

The related changes of the orbital averaged potential have been calculated for the coupled phonon-plasmon modes. In particular for the phonon-like mode we have

found a very strong enhancement in the nonadiabatic sector of phase space as compared with the adiabatic result. The possible importance of the phonon-like O_z^Z mode for the electron self-energy has been pointed out. This demonstrates again the importance of the long-ranged polar coupling of the electrons and the c -axis phonons. As far as the low energy scale and the bosonic glue to keep the electron pairs in the cuprates together is con-

cerned our calculations make a strong case that phonon-plasmon modes from the nonadiabatic zone around the c -axis have to be added to the list of possible players. Finally, it has been speculated within the phonon-plasmon scenario why SrRuO could be a low-temperature superconductor in contrast to the high-temperature superconductor LaCuO.

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